MAELAS: MAgneto-ELAStic properties calculation via computational high-throughput approach

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Abstract

In this work, we present the program MAELAS to calculate anisotropic magnetostrictive coefficients and magnetoelastic constants in an automated way by Density Functional Theory calculations. To illustrate the methodology of MAELAS, we present a review of the theoretical background of magnetostriction for the main crystal symmetries in this field that are implemented in the code. As a benchmark, some tests are shown for well-known magnetic materials.

Keywords: Magnetostriction, Magnetoelasticity, High-throughput computation, First-principles calculations

PROGRAM SUMMARY

Program Title: MAELAS
Developer’s repository link: https://github.com/pnieves2019/MAELAS
Licensing provisions: BSD 3-clause
Programming language: Python3
Nature of problem: To calculate anisotropic magnetostrictive coefficients and magnetoelastic constants in an automated way based on Density Functional Theory methods.
Solution method: After a crystal symmetry analysis, a set of distorted lattice and spin configurations are generated using the pymatgen library [1]. Next, the energy of these states

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is calculated by the first-principles code VASP [2], including the spin-orbit coupling. The magnetostrictive coefficients are derived from the fitting of these energies to a quadratic function [3]. Finally, if the elastic tensor is provided [4], then the magnetoelastic constants are calculated too.

Additional comments including restrictions and unusual features: This version supports the following crystal systems: Cubic (point groups 432, 4\(\bar{3}\)m, \(\bar{3}m\bar{3}\)), Hexagonal (6\(mm\), 6\(2m\), 6/\(mmm\)), Trigonal (32, 3\(m\), \(\bar{3}m\)), Tetragonal (4\(mm\), 4\(22\), 4\(\bar{2}m\), 4/\(mmm\)), and Orthorhombic (2\(22\), 2\(mm\), \(mmm\)).

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1. Introduction

A magnetostrictive material is one which changes in size due to a change of state of magnetization. These materials are characterized by magnetostrictive coefficients (\(\lambda\)). In many technical applications such as electric transformers, motor shielding, and magnetic recording, magnetic materials with extremely small magnetostrictive coefficients are required. By contrast, materials with large magnetostrictive coefficients are needed for many applications in electromagnetic microdevices as actuators and sensors [1]. Typically, elementary Rare-Earth (R) metals (under low temperature and high magnetic field) and compounds with R and transition metals exhibit a high magnetostriction (\(\lambda > 10^{-3}\)). In particular, the highest magnetostrictions were found in the RFe\(_2\) compounds with Laves phase C15 structure type (face centered cubic) [2]. For instance, Terfenol-D (Tb\(_{0.27}\)Dy\(_{0.73}\)Fe\(_2\)) is a widely used magnetostrictive material thanks to its giant magnetostriction along [111] crystallographic direction (\(\lambda_{111} = 1.6 \times 10^{-3}\)) under moderate magnetic fields (< 2 kOe) at room temperature [3]. Beyond cubic systems, the research of magnetostrictive materials has been focused on hexagonal
crystals like RCo$_5$ (space group 191), hexagonal and trigonal R$_2$Co$_7$ and R$_2$Co$_{17}$ series, and tetragonal R$_2$Fe$_{14}$B [4, 5]. More recently, the problem of R availability [6] has also motivated the exploration of R-free magnetostrictive materials like Galfenol (Fe-Ga) or spinel ferrites (CoFe$_2$O$_4$).

Concerning the theory of magnetostriction, the basic equations for cubic (I) crystals were developed by Akulov [7] and Becker et al. [8] in the 1920s and 30s. In the next three decades, great advances took place due to the outstanding works of Mason [9], Clark et al. [10], and Callen and Callen [11], as well as many others, where the theory was extended to other crystal symmetries. Over the last decades, modern electronic structure theory based on Density Functional Theory (DFT) has been successfully applied to describe magnetostriction of many materials [11, 12-24]. Nowadays, a common method to calculate magnetostrictive coefficients is to deduce them from the energy of a set of distorted unit cells with specific spin directions [12, 13]. In this work, we present the MAELAS program where this methodology is implemented and generalized for the main crystal symmetries in the research field of magnetostriction. In Section 2, we review some theoretical concepts and equations of magnetostriction, on which MAELAS program is based. In Section 3, we explain in detail the methodology and workflow of the program, while some examples are shown in Section 4. Finally, the paper ends with a summary of the main conclusions and future perspectives (Section 5).

2. Theory of magnetostriction

The magnetostrictive response is mainly originated by two kind of sources: (i) isotropic exchange interaction (volume strain) and (ii) spin-orbit coupling (anisotropic strain) [5]. The magnetostriction due to isotropic exchange leads to fractional volume changes, so that it doesn’t depend on the magnetization direction [25]. On the other hand, the spin-orbit coupling is responsible for the magnetostriction that depends on the magnetization orientation (anisotropic). The current version of the program MAELAS calculates the magnetostrictive coefficients and magnetoelastic constants related to the anisotropic magnetostriction.

Let’s consider $l_0$ the initial length of a demagnetized material along the direction $\hat{\beta}$ ($|\hat{\beta}| = 1$), and $l$ the final length along the same direction $\hat{\beta}$ when the system is magnetized along the direction $\hat{\alpha}$ ($|\hat{\alpha}| = 1$). The relative length change $(l - l_0)/l_0 = \Delta l/l_0$ can be written as [5]

$$\frac{\Delta l}{l_0} \bigg|_{\hat{\alpha} \rightarrow \hat{\beta}} = \sum_{i,j=x,y,z} \varepsilon_{ij}^{eq}(\hat{\alpha}) \hat{\beta}_i \hat{\beta}_j,$$

(1)
Figure 1: Magnetostriction of a single crystal under an external magnetic field ($\alpha \parallel H$) perpendicular to the measured length direction ($\beta \perp H$). Symbols $M$ and $M_s$ stand for macroscopic magnetization and saturation magnetization, respectively. Dash line on the right represents the original size of the demagnetized material. The magnetostriction effect has been magnified in order to help to visualize it easily, in real materials it is smaller ($\Delta l/l_0 \sim 10^{-3} \sim 10^{-6}$).

where $\varepsilon_{eq}^{ij}$ is the equilibrium strain tensor. Fig.1 shows a sketch of magnetostriction. The Eq.1 is the governing equation of magnetostriction, once it is rewritten in terms of the magnetostrictive coefficients ($\lambda$) conveniently. To do so, one needs to derive $\varepsilon_{eq}^{ij}$ by minimizing the elastic ($E_{el}$) and magnetoelastic ($E_{me}$) energies

$$\frac{\partial (E_{el} + E_{me})}{\partial \varepsilon_{ij}} = 0.$$  \hspace{1cm} (2)

The total energy must be invariant under the symmetry operations of the crystal lattice [2]. The elastic energy up to second-order is given by

$$E_{el} = E_0 + \frac{V_0}{2} \sum_{i,j=1}^{6} c_{ij} \varepsilon_i \varepsilon_j + O(\varepsilon^3),$$  \hspace{1cm} (3)

where $E_0$ and $V_0$ are the equilibrium energy and volume, and $c_{ij}$ is the elastic tensor. Here, we used the standard Voigt notation for the indices of strains and elastic tensors, contracting a pair of Cartesian indices into a single integer: $xx \to 1$, $yy \to 2$, $zz \to 3$, $yz \to 4$, $xz \to 5$, $xy \to 6$. The magnetoelastic energy $E_{me}$ comes from the strain dependence of the magnetocrystalline anisotropy energy (MAE).
$E_K$ \[26\]

\[
E_K = E_K^0 + \sum_{i=1}^{6} \frac{\partial E_K^0}{\partial \varepsilon_i} \varepsilon_i + \frac{1}{2} \sum_{i,j=1}^{6} \frac{\partial^2 E_K^0}{\partial \varepsilon_i \partial \varepsilon_j} \varepsilon_i \varepsilon_j + \ldots,
\]

\[(4)\]

where $E_K^0$ corresponds to the MAE of the undeformed state that contains the magnetocrystalline anisotropy constants $K$. The third term in the right hand side of Eq\[4\] is the second-order magnetoelastic energy that leads to a very small additional contribution to the second-order elastic energy given by Eq\[3\] which is, usually, neglected \[26, 27\]. The first-order magnetoelastic energy

\[
E_{me} = \sum_{i=1}^{6} \frac{\partial E_K^0}{\partial \varepsilon_i} \varepsilon_i
\]

\[(5)\]

is obtained by taking the direct product of the symmetry strains and direction cosine polynomial for each irreducible representation, multiplying by a constant, called the magnetoelastic constant and finally summing over the different representations \[2, 5, 10, 11\]. Frequently, the first-order magnetoelastic energy is considered up to second-order of the direction cosine polynomial. In cartesian coordinates, it may be written as

\[
E_{me} = \sum_{i=1}^{3} g_i(\alpha^0) \varepsilon_i + \sum_{i=1}^{6} f_i(\alpha^2) \varepsilon_i + O(\alpha^4),
\]

\[(6)\]

where functions $g_i$ and $f_i$ contain the magnetoelastic constants ($b$). In the following, we show the form of Eqs\[3, 3\] and \[6\] for the main crystal symmetries studied in magnetostriction, which are implemented in the program MAELAS. The remaining crystal systems not discussed here might be included in the new versions of the code. In Table\[1\] we present a summary of the crystal systems supported by MAELAS. Here, we use the notation of Wallace \[28\] (I/II) to distinguish Laue classes within the same crystal system.

2.1. Cubic (I)

For cubic (I) systems (point groups 432, 43$m$, m3$m$), there are three independent elastic constants $c_{11}$, $c_{12}$ and $c_{44}$, so Eq\[3\] becomes

\[
E_{el}^{cub} = \frac{c_{11}}{2}(\varepsilon_{xx}^2 + \varepsilon_{yy}^2 + \varepsilon_{zz}^2) + c_{12}(\varepsilon_{xx} \varepsilon_{yy} + \varepsilon_{xx} \varepsilon_{zz} + \varepsilon_{yy} \varepsilon_{zz}) + \frac{c_{44}}{2}(\varepsilon_{xy}^2 + \varepsilon_{yz}^2 + \varepsilon_{xz}^2).
\]

\[(7)\]

On the other hand, the first-order magnetoelastic energy up to second-order direction cosine polynomial contains 3 magnetoelastic constants \[11\]. From the
Table 1: Number of independent second-order elastic constants of each crystal system. Number of independent magnetoelastic and magnetostrictive coefficients up to second-order of the direction cosine polynomial in the first-order magnetoelastic energy. In the last column we specify which crystal systems are supported by the current version of MAELAS.

| Crystal system | Point groups | Space groups | Elastic constants \((c_{ij})\) | Magnetoelastic constants \((b)\) | Magnetostriction coefficients \((\lambda)\) | MAELAS |
|----------------|--------------|--------------|-------------------------------|-------------------------------|---------------------------------|--------|
| Triclinic      | 1, 1         | 1–2          | 21                            | 36                            | 36                              | No     |
| Monoclinic     | 2, 2/m       | 3–15         | 13                            | 20                            | 20                              | No     |
| Orthorhombic   | 222, 2mm,mmm | 16–74        | 9                             | 12                            | 12                              | Yes    |
| Tetragonal (II)| 4, 4, 4/m    | 75–88        | 7                             | 10                            | 10                              | No     |
| Tetragonal (I) | 4mm, 42m, 4/mnm | 89–142 | 6                             | 7                             | 7                               | Yes    |
| Trigonal (II)  | 3, 3         | 143–148      | 7                             | 12                            | 12                              | No     |
| Trigonal (I)   | 32, 3m, 3m   | 149–167      | 6                             | 8                             | 8                               | Yes    |
| Hexagonal (II) | 6, 6, 6/m    | 168–176      | 5                             | 8                             | 8                               | No     |
| Hexagonal (I)  | 6mm, 62m, 6/mmnm | 177–194 | 5                             | 6                             | 6                               | Yes    |
| Cubic (II)     | 23, m3       | 195–206      | 3                             | 4                             | 4                               | No     |
| Cubic (I)      | 432, 43m, m3m | 207–230     | 3                             | 3                             | 3                               | Yes    |

Inserting these equilibrium strains into Eq.\(1\) gives
\[
\frac{\Delta l}{l_0} = \lambda^\alpha + 3\lambda^\beta\left(\alpha_x^2\beta_x^2 + \alpha_y^2\beta_y^2 + \alpha_z^2\beta_z^2 - \frac{1}{3}\right) + 3\lambda^\gamma(\alpha_x\alpha_y\beta_x + \alpha_y\alpha_z\beta_y + \alpha_z\alpha_x\beta_z),
\]
where
\[ \lambda^\alpha = -b_0 - \frac{1}{3}b_1 \]
\[ \lambda_{001} = \frac{-2b_1}{3(c_{11} - c_{12})} \]
\[ \lambda_{111} = -\frac{2}{3}c_{44} \]

The coefficient \( \lambda^\alpha \) describes the volume magnetostriction, while \( \lambda_{001} \) and \( \lambda_{111} \) are the anisotropic magnetostrictive coefficients that give the fractional length change along the [001] and [111] directions when a demagnetized material is magnetized in these directions, respectively. The superscript \( \alpha \) in \( \lambda^\alpha \) stands for one irreducible representation of the group of transformations which take the crystal into itself \([2, 5]\), so it should not be confused with the direction of magnetization \( \alpha \). The theory of magnetostriction for polycrystalline materials is more complex. A widely used approximation is to assume that the stress distribution is uniform through the material. In this case the relative change in length may be put into the form \([5, 7, 26, 29]\)

\[ \frac{\Delta l}{l_0} \bigg|_{\beta} = \frac{3}{2} \lambda_S \left[ (\alpha \cdot \beta)^2 - \frac{1}{3} \right] \]

where
\[ \lambda_S = \frac{2}{5} \lambda_{001} + \frac{3}{5} \lambda_{111} \]

This result is analogous to the Reuss approximation used in the elastic theory of polycrystals to obtain a lower bound of bulk and shear modulus \([5, 30-32]\).

2.2. Hexagonal (I)

The hexagonal (I) system (point groups 6mm, 622, \( \bar{6}2m \), 6/\( mmm \)) has five independent elastic constants \( c_{11}, c_{12}, c_{13}, c_{33} \) and \( c_{44} \), where \( c_{66} = (c_{11} - c_{12})/2 \). Thus, Eq.[3] reads

\[ E_{el}^{hex} = \frac{1}{2} c_{11}(\varepsilon_{xx}^2 + \varepsilon_{yy}^2) + c_{12}\varepsilon_{xx}\varepsilon_{yy} + c_{13}(\varepsilon_{xx} + \varepsilon_{yy})\varepsilon_{zz} + \frac{1}{2} c_{33}\varepsilon_{zz}^2 + \frac{1}{2} c_{44}(\varepsilon_{yz}^2 + \varepsilon_{xz}^2) + \frac{1}{4}(c_{11} - c_{12})\varepsilon_{xy}^2 \]

The first order magnetoelastic energy up to quadratic direction cosine polynomial contains 6 magnetoelastic constants \([11]\). In cartesian coordinates it can be written
as \[2, 5\]

\[E_{\text{hex}(f)} = b_{11}(\varepsilon_{xx} + \varepsilon_{yy}) + b_{12}\varepsilon_{zz} + b_{21}\left(\varepsilon_{xx} - \frac{1}{3}\right)(\varepsilon_{xx} + \varepsilon_{yy}) + b_{22}\left(\varepsilon_{zz} - \frac{1}{3}\right)\varepsilon_{zz} + \frac{1}{2}b_3[(\alpha_x - \alpha_y)(\varepsilon_{xx} - \varepsilon_{yy}) + 2\alpha_x\alpha_y\varepsilon_{xy}] + b_4(\alpha_x\alpha_z\varepsilon_{xz} + \alpha_y\alpha_z\varepsilon_{yz}).\]  

(15)

Once the equilibrium strains are calculated by minimizing Eqs. 14 and 15 through Eq. 2 and inserted into Eq. 1, one finds \[2, 5, 10\]

\[\Delta \varepsilon_l|_{l_0}^\alpha = \lambda^{\alpha 1.0}(\beta_x^2 + \beta_y^2) + \lambda^{\alpha 2.0}\beta_z^2 + \lambda^{\alpha 1.2}\left(\alpha_z^2 - \frac{1}{3}\right)(\beta_x^2 + \beta_y^2) + \lambda^{\alpha 2.2}\left(\alpha_z^2 - \frac{1}{3}\right)\beta_z^2 + \lambda^{\gamma 2}\left[\frac{1}{2}(\alpha_x^2 - \alpha_y^2)(\beta_x^2 - \beta_y^2) + 2\alpha_x\alpha_y\beta_x\beta_y\right] + 2\lambda^{\varepsilon 2}(\alpha_x\alpha_z\beta_x\beta_z + \alpha_y\alpha_z\beta_x\beta_z),\]  

(16)

where

\[\lambda^{\alpha 1.0} = \frac{b_{11}c_{33} + b_{12}c_{13}}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2},\]
\[\lambda^{\alpha 2.0} = \frac{2b_{11}c_{13} - b_{12}(c_{11} + c_{12})}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2},\]
\[\lambda^{\alpha 1.2} = \frac{-b_{21}c_{33} + b_{22}c_{13}}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2},\]
\[\lambda^{\alpha 2.2} = \frac{-b_{21}c_{13} - b_{22}(c_{11} + c_{12})}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2},\]
\[\lambda^{\gamma 2} = \frac{-b_3}{c_{11} - c_{12}},\]
\[\lambda^{\varepsilon 2} = \frac{-b_4}{2c_{44}}.\]  

(17)

These magnetostrictive coefficients are related to the normal strain modes for a cylinder \[2, 5\]. In literature there are different arrangements of the right hand side of Eq. 16 that leads to other definitions of the magnetostrictive coefficients, like those defined by Mason \[9\], Birss \[26\], and Callen and Callen \[11\]. The conversion formulas between Eq. 16 and all these other conventions can be found in Appendix A. These conversion formulas are implemented in the program MAE-LAS, so that the magnetostrictive coefficients are also given according to these definitions.
2.3. Trigonal (I)

The trigonal (I) system (point groups $32$, $3m$, $\bar{3}m$) has 6 independent elastic constants $c_{11}$, $c_{12}$, $c_{13}$, $c_{33}$, $c_{44}$ and $c_{14}$, where $c_{66} = (c_{11} - c_{12})/2$. The elastic energy reads

$$E_{el}^{\text{trig}(I)} = \frac{1}{2} c_{11} \varepsilon_{xx}^2 + c_{12} \varepsilon_{xx} \varepsilon_{yy} + c_{13} (\varepsilon_{xx} + \varepsilon_{yy}) \varepsilon_{zz} + \frac{1}{2} c_{33} \varepsilon_{zz}^2$$
$$+ \frac{1}{2} c_{44} (\varepsilon_{xz}^2 + \varepsilon_{yz}^2) + \frac{1}{4} (c_{11} - c_{12}) \varepsilon_{xy}^2 + c_{14} (\varepsilon_{xy} \varepsilon_{xz} + \varepsilon_{xx} \varepsilon_{yz} - \varepsilon_{yy} \varepsilon_{yz}).$$

(18)

The magnetoelastic energy contains 8 magnetoelastic constants \[11\]. In cartesian coordinates it can be written as \[5\]

$$E_{me}^{\text{trig}(I)} = b_{11} (\varepsilon_{xx} + \varepsilon_{yy}) + b_{12} \varepsilon_{zz} + b_{21} \left( \alpha_x^2 - \frac{1}{3} \right) (\varepsilon_{xx} + \varepsilon_{yy}) + b_{22} \left( \alpha_y^2 - \frac{1}{3} \right) \varepsilon_{zz}$$
$$+ \frac{1}{2} b_3 [(\alpha_x^2 - \alpha_y^2) (\varepsilon_{xx} - \varepsilon_{yy}) + 2 \alpha_x \alpha_y] + b_4 (\alpha_x \alpha_z \varepsilon_{xz} + \alpha_y \alpha_z \varepsilon_{yz})$$
$$+ b_{14} \left[ \frac{1}{2} (\alpha_x^2 - \alpha_y^2) \varepsilon_{yz} + \alpha_x \alpha_z \varepsilon_{xz} \right] + b_{34} \left[ \frac{1}{2} \alpha_x \alpha_z (\varepsilon_{xx} - \varepsilon_{yy}) + \alpha_x \alpha_z \varepsilon_{xy} \right].$$

(19)

Next, we obtain the equilibrium strains via Eq.2. Replacing them into Eq.1 leads to \[5\]

$$\frac{\Delta l}{l_0} \bigg|_{\alpha \beta} = \lambda^{\alpha 1.0} (\beta_x^2 + \beta_y^2) + \lambda^{\alpha 2.0} \beta_z^2 + \lambda^{\alpha 1.2} \left( \alpha_z^2 - \frac{1}{3} \right) (\beta_x^2 + \beta_y^2)$$
$$+ \lambda^{\alpha 2.2} \left( \alpha_z^2 - \frac{1}{3} \right) \beta_z^2 + \lambda^{\alpha 1.1} \left[ \frac{1}{2} (\alpha_x^2 - \alpha_y^2) (\beta_x^2 - \beta_y^2) + \alpha_x \alpha_y \beta_x \beta_y \right]$$
$$+ \lambda^{\gamma 2} (\alpha_x \alpha_z \beta_x \beta_z + \alpha_y \alpha_z \beta_x \beta_z) + \lambda_{12} \left[ \frac{1}{2} \alpha_x \alpha_z (\beta_x^2 - \beta_y^2) + \alpha_x \alpha_z \beta_x \beta_y \right]$$
$$+ \lambda_{21} \left[ \frac{1}{2} (\alpha_x^2 - \alpha_y^2) \beta_y \beta_z + \alpha_x \alpha_z \beta_x \beta_z \right],$$

(20)
where

\[ \lambda_{\alpha,0}^{1} = \frac{b_{11}c_{33} + b_{12}c_{13}}{c_{33}(c_{11} + c_{12}) - 2c_{13}^{2}} \],
\[ \lambda_{\alpha,2}^{0} = \frac{2b_{11}c_{13} - b_{12}(c_{11} + c_{12})}{c_{33}(c_{11} + c_{12}) - 2c_{13}^{2}} \],
\[ \lambda_{\alpha,1,2} = \frac{-b_{21}c_{33} + b_{22}c_{13}}{c_{33}(c_{11} + c_{12}) - 2c_{13}^{2}} \],
\[ \lambda_{\alpha,2,2}^{1} = \frac{2b_{21}c_{13} - b_{22}(c_{11} + c_{12})}{c_{33}(c_{11} + c_{12}) - 2c_{13}^{2}} \],
\[ \lambda_{\gamma,1} = \frac{c_{14}b_{14} - c_{44}b_{3}}{2c_{44}(c_{11} - c_{12}) - c_{14}^{2}} \],
\[ \lambda_{\gamma,2} = \frac{1}{2}b_{4}(c_{11} - c_{12}) - b_{34}c_{14} \]
\[ \frac{1}{2}c_{44}(c_{11} - c_{12}) - c_{14}^{2} \],
\[ \lambda_{12} = \frac{c_{14}b_{4} - c_{44}b_{34}}{2c_{44}(c_{11} - c_{12}) - c_{14}^{2}} \]
\[ \lambda_{21} = \frac{1}{2}b_{14}(c_{11} - c_{12}) - b_{34}c_{14} \]
\[ \frac{1}{2}c_{44}(c_{11} - c_{12}) - c_{14}^{2} \].

(21)

2.4. Tetragonal (I)

The tetragonal (I) crystal system (point groups 4\textit{mm}, 422, \bar{4}2m, 4/\textit{mmm}) has six independent elastic constants \( c_{11}, c_{12}, c_{13}, c_{33}, c_{44} \) and \( c_{66} \). The elastic energy is given by

\[
E_{\text{el}}^{\text{tet}} = \frac{1}{2}c_{11}(\varepsilon_{xx}^{2} + \varepsilon_{yy}^{2}) + c_{12}\varepsilon_{xx}\varepsilon_{yy} + c_{13}(\varepsilon_{xx} + \varepsilon_{yy})\varepsilon_{zz} + \frac{1}{2}c_{33}\varepsilon_{zz}^{2} \]
\[ + \frac{1}{2}c_{44}(\varepsilon_{xz}^{2} + \varepsilon_{yz}^{2}) + \frac{1}{2}c_{66}\varepsilon_{xy}^{2} \].

(22)

On the other hand, there are 7 independent magnetoelastic constants \([11]\). The magnetoelastic energy can be written as \([5][24]\)

\[
E_{\text{me}}^{\text{tet}(I)} = b_{11}(\varepsilon_{xx} + \varepsilon_{yy}) + b_{12}\varepsilon_{zz} + b_{21}\left(\alpha_{x}^{2} - \frac{1}{3}\right)\varepsilon_{xx} + b_{22}\left(\alpha_{y}^{2} - \frac{1}{3}\right)\varepsilon_{yy} + \frac{1}{2}b_{3}(\alpha_{x}^{2} - \alpha_{y}^{2})\varepsilon_{xx} - \varepsilon_{xy} + b_{3}'\alpha_{x}\alpha_{y}\varepsilon_{xy} + b_{4}(\alpha_{x}\alpha_{y}\varepsilon_{xx} + \alpha_{y}\alpha_{z}\varepsilon_{yz}) \]

(23)
After the equilibrium strains are calculated by minimizing Eqs. 22 and 23 through Eq. 2 and replaced into Eq. 1, we have

\[ \Delta l = \left| \lambda^{\alpha 1.0} (\beta_x^2 + \beta_y^2) + \lambda^{\alpha 2.0} \beta_z^2 + \lambda^{\alpha 1.2} \left( \alpha_x^2 - \frac{1}{3} \right) (\beta_x^2 + \beta_y^2) \right| \]

\[ + \lambda^{\alpha 2.2} \left( \alpha_x^2 - \frac{1}{3} \right) \beta_z^2 + \frac{1}{2} \lambda^{\gamma 2} (\alpha_x^2 - \alpha_y^2) (\beta_x^2 - \beta_y^2) + 2 \lambda^{\delta 2} \alpha_x \alpha_y \beta_x \beta_y \]

\[ + 2 \lambda^{\epsilon 2} (\alpha_x \alpha_z \beta_x + \alpha_y \alpha_z \beta_y), \]

where

\[ \lambda^{\alpha 1.0} = \frac{b_{11} c_{33} + b_{12} c_{13}}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2}, \]

\[ \lambda^{\alpha 2.0} = \frac{2b_{11} c_{13} - b_{12}(c_{11} + c_{12})}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2}, \]

\[ \lambda^{\alpha 1.2} = \frac{-b_{21} c_{33} + b_{22} c_{13}}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2}, \]

\[ \lambda^{\alpha 2.2} = \frac{2b_{21} c_{13} - b_{22}(c_{11} + c_{12})}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2}, \]

\[ \lambda^{\gamma 2} = \frac{-b_3}{c_{11} - c_{12}}, \]

\[ \lambda^{\delta 2} = \frac{-b_3'}{2c_{66}}, \]

\[ \lambda^{\epsilon 2} = \frac{-b_4}{2c_{44}}. \]

Mason derived an equivalent equation to Eq. 24 using a different arrangement of the terms and definitions of the magnetostrictive coefficients. The conversion formulas between the magnetostrictive coefficients in Eq. 24 and those defined by Mason are shown in Appendix B. These conversion formulas are implemented in the program MAELAS, so that the magnetostrictive coefficients are given using both definitions.

2.5. Orthorhombic

The orthorhombic crystal system (point groups 222, 2mm, mmm) has 9 independent elastic constants \( c_{11}, c_{12}, c_{13}, c_{22}, c_{23}, c_{33}, c_{44}, c_{55} \) and \( c_{66} \). The elastic
energy is given by

\[
E_{\text{el}} \propto \frac{1}{2} c_{11} \varepsilon_{xx}^2 + \frac{1}{2} c_{22} \varepsilon_{yy}^2 + c_{12} \varepsilon_{xx} \varepsilon_{yy} + c_{13} \varepsilon_{xx} \varepsilon_{zz} + c_{23} \varepsilon_{yy} \varepsilon_{zz} + \frac{1}{2} c_{33} \varepsilon_{zz}^2
\]

\[
+ \frac{1}{2} c_{44} \varepsilon_{xz}^2 + \frac{1}{2} c_{55} \varepsilon_{yz}^2 + \frac{1}{2} c_{66} \varepsilon_{xy}^2.
\]

(26)

The magnetoelastic energy contains 12 independent magnetoelastic constants [11]. Mason derived the following expression of the relative length change [9]

\[
\frac{\Delta l}{l_0} \bigg|_\alpha = \lambda \alpha^1.0 \beta_x^2 + \lambda \alpha^2.0 \beta_y^2 + \lambda \alpha^3.0 \beta_z^2 + \lambda_1 (\alpha_x \beta_x \beta_y - \alpha_x \alpha_y \beta_x \beta_y)
\]

\[
+ \lambda_2 (\alpha_x \beta_x^2 - \alpha_x \alpha_y \beta_x \beta_y) + \lambda_3 (\alpha_x \beta_y^2 - \alpha_x \alpha_y \beta_x \beta_y)
\]

\[
+ \lambda_4 (\alpha_x \beta_z^2 - \alpha_x \alpha_y \beta_x \beta_y - \alpha_y \alpha_z \beta_x \beta_y) + \lambda_5 (\alpha_x \beta_z^2 - \alpha_x \alpha_y \beta_x \beta_y)
\]

\[
+ \lambda_6 (\alpha_x \beta_z^2 - \alpha_x \alpha_y \beta_x \beta_y) + 4 \lambda_7 \alpha_x \alpha_y \beta_x \beta_y + 4 \lambda_8 \alpha_x \alpha_y \beta_x \beta_y + 4 \lambda_9 \alpha_x \alpha_y \beta_x \beta_y.
\]

(27)

Note that we added the terms that describes the volume magnetostriction (\(\lambda \alpha^{1.0}\), \(\lambda \alpha^{2.0}\), and \(\lambda \alpha^{3.0}\)), which were not included in the original work of Mason [9]. Additionally, the expression of the magnetoelastic energy and the relations between magnetostrictive coefficients, elastic and magnetoelastic constants were not shown by Mason either. For completeness, here we deduce it from Eqs. [26] and [27]. To do so, we aim to find the unknown functions \(g_i\) and \(f_i\) in the general form of the magnetoelastic energy in cartesian coordinates given by Eq. [6]. Firstly, we minimize Eqs. [26] and [6] via Eq. [2]. This gives a set of equations that links the unknown functions \(g_i\) and \(f_i\) with the equilibrium strains. Next, we extract the equilibrium strains by direct comparison between Eqs. [1] and [27]. Finally, we substitute the equilibrium strains into the set of equations that relates \(g_i\) and \(f_i\) with the equilibrium strains, from which we obtain \(g_i\) and \(f_i\). Inserting the calculated \(g_i\) and \(f_i\) into Eq. [6] we have

\[
E_{\text{me}} = b_{01} \varepsilon_{xx} + b_{02} \varepsilon_{yy} + b_{03} \varepsilon_{zz} + b_1 \alpha_x^2 \varepsilon_{xx} + b_2 \alpha_y^2 \varepsilon_{yy} + b_3 \alpha_z^2 \varepsilon_{zz} + b_4 \alpha_x^2 \varepsilon_{xy} + b_5 \alpha_x^2 \varepsilon_{xz}
\]

\[
+ b_6 \alpha_x^2 \varepsilon_{yz} + b_7 \alpha_y \alpha_x \varepsilon_{xx} + b_8 \alpha_x \alpha_y \varepsilon_{xx} + b_9 \alpha_y \alpha_x \varepsilon_{xx}.
\]

(28)
where

\[ b_{01} = -c_{11} \lambda_{\alpha_1} - c_{12} \lambda_{\alpha_2} - c_{13} \lambda_{\alpha_3} \]
\[ b_{02} = -c_{12} \lambda_{\alpha_1} - c_{22} \lambda_{\alpha_2} - c_{23} \lambda_{\alpha_3} \]
\[ b_{03} = -c_{13} \lambda_{\alpha_1} - c_{23} \lambda_{\alpha_2} - c_{33} \lambda_{\alpha_3} \]
\[ b_1 = -c_{11} \lambda_1 - c_{12} \lambda_3 - c_{13} \lambda_5 \]
\[ b_2 = -c_{11} \lambda_2 - c_{12} \lambda_4 - c_{13} \lambda_6 \]
\[ b_3 = -c_{12} \lambda_1 - c_{22} \lambda_3 - c_{23} \lambda_5 \]
\[ b_4 = -c_{12} \lambda_2 - c_{22} \lambda_4 - c_{23} \lambda_6 \]
\[ b_5 = -c_{13} \lambda_1 - c_{23} \lambda_3 - c_{33} \lambda_5 \]
\[ b_6 = -c_{13} \lambda_2 - c_{23} \lambda_4 - c_{33} \lambda_6 \]
\[ b_7 = c_{66} (\lambda_1 + \lambda_2 + \lambda_3 + \lambda_4 - 4 \lambda_7) \]
\[ b_8 = c_{55} (\lambda_1 + \lambda_5 - 4 \lambda_8) \]
\[ b_9 = c_{44} (\lambda_4 + \lambda_6 - 4 \lambda_9) \]

(29)

Alternatively, one can deduce the magnetoelastic energy using the general approach based on the symmetry strains and direction cosine polynomial for each irreducible representation [2, 5, 11]. This approach may lead to different definitions of the elastic constants and magnetostrictive coefficients, as we have discussed for the hexagonal (I) and tetragonal (I) systems in Appendix A and Appendix B respectively. A generalization of the approach taken by Becker and Doring [8] for orthorhombic crystals can be found in Ref. [33].

3. Methodology

3.1. Calculation of magnetostrictive coefficients and magnetoelastic constants

The methodology implemented in the program MAELAS to calculate the anisotropic magnetostrictive coefficients is a generalization of the approach proposed by Wu and Freeman for cubic crystals [12, 13]. Namely, for each magnetostrictive coefficient (\( \lambda^i \)) we select one measurement length direction \( \beta^i \) and two magnetization directions \( \alpha_1^i \) and \( \alpha_2^i \) in such a way that we have

\[
\frac{\Delta l}{l_0} \bigg|_{\beta^i} \alpha_1^i - \frac{\Delta l}{l_0} \bigg|_{\beta^i} \alpha_2^i = \eta^i \lambda^i,
\]

(30)
where $\eta^i_1$ is a real number. In Table 2 we show the selected set of $\beta^i_1$, $\alpha^i_1$, and $\alpha^i_2$ that fulfils Eq.30 for each $\lambda^i$. Next, the left hand side of Eq.30 is written as

$$\Delta l_{\beta} |_{\alpha^i_1} - \Delta l_{\beta} |_{\alpha^i_2} = \frac{l_1 - l_0}{l_0} - \frac{l_2 - l_0}{l_0} = \frac{2(l_1 - l_2)}{(l_1 + l_2)} \left[ 1 - \frac{l_1 + l_2 - 2l_0}{l_1 + l_2} \right]$$

(31)

where the last approximation is based on the fact that $|l_1(2) - l_0|/l_0 < 10^{-2}$. For instance, a very large value of $\Delta l/l_0$ is about $4.5 \times 10^{-3}$ found in TbFe$_2$ (Laves phase C15) along direction [111] at $T = 0K$. This approximation allows us to skip $l_0$ (length along $\beta$ in the macroscopic demagnetized state) which can’t be calculated with DFT methods. The quantities $l_1$ and $l_2$ correspond to the cell length along $\beta$ when the magnetization points to $\alpha_1$ and $\alpha_2$, respectively, and we can calculate them in the following way. Firstly, we generate a set of volume-conserving distorted unit cells applying different strain values along $\beta$, see Appendix C. For each distorted cell, we calculate the energy constraining the spins to the directions given by $\alpha_1$ and $\alpha_2$. Next, the energy versus the cell length along $\beta$ for each spin direction $\alpha_{1(2)}$ is fitted to a quadratic function

$$E(l) |_{\beta} = A_j l^2 + B_j l + C_j, \quad j = 1, 2.$$  

(32)

The minimum of this quadratic function for spin direction $\alpha_{1(2)}$ corresponds to $l_{1(2)} = -B_{1(2)}/(2A_{1(2)})$. Once we have $l_1$ and $l_2$, we can obtain the magnetostrictive coefficients combining Eqs.30 and 31, that is,

$$\lambda^i = \frac{2(l_1 - l_2)}{\eta^i(l_1 + l_2)},$$

(33)

where the value of $\eta^i$ for each $\lambda^i$ is given in Table 2. In Table 2 we see that our choice of $\beta$ and $\alpha_{1(2)}$ makes $\eta$ depend on some magnetostrictive coefficients for $\lambda_7$, $\lambda_8$ and $\lambda_9$ in orthorhombic crystals. For instance, $\lambda_7$ reads

$$\lambda_7 = \frac{(a^2 + b^2)(l_1 - l_2)}{ab(l_1 + l_2)} - \frac{(a-b)(a[\lambda_4 + \lambda_2] - b[\lambda_3 + \lambda_4])}{4ab},$$

(34)

where $a$ and $b$ are the relaxed (not distorted) lattice parameters of the unit cell. Here, we need to use the values of $\lambda_1$, $\lambda_2$, $\lambda_3$ and $\lambda_4$ calculated previously in order
to obtain $\lambda_7$. Note that a simpler expression for $\lambda_7$ can be achieved choosing the length measurement direction $\mathbf{\beta} = \left( \frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0 \right)$. However, from a computational point of view, it is easier to extract the cell length $l$ along $\mathbf{\beta} = \left( \frac{a}{\sqrt{a^2+b^2}}, \frac{b}{\sqrt{a^2+b^2}}, 0 \right)$ of each distorted cell. Similarly, one can easily deduce the explicit equation for $\lambda_8$ and $\lambda_9$. The magnetostrictive coefficients can be written as [14]

$$\lambda_i \approx -\frac{1}{\eta_i B_1} \cdot \frac{\partial}{\partial l} \left[ \frac{\alpha_2}{\mathbf{\beta}} - \frac{\alpha_1}{\mathbf{\beta}} \right] (l = l_2), \quad (35)$$

where $B_1$ is always negative. Hence, we see that the sign and strength of each magnetostrictive coefficient is determined by the derivative of the energy difference between states with spin directions $\alpha_2$ and $\alpha_1$ with respect to the cell length along $\mathbf{\beta}$ evaluated at $l = l_2$. Note that the analysis of Eq.35 for $\lambda_7$, $\lambda_8$ and $\lambda_9$ in orthorhombic crystals is more complicated than in the other cases due to the dependence of $\eta$ on some magnetostrictive coefficients.

Finally, if the elastic constants ($c_{nm}$) are provided as inputs, then the magnetoelastic constants ($b_k$) are directly deduced from the relations between them, $b_k = b_k(\lambda_i, c_{nm})$, given in Section 2 for each crystal system (Eqs 11, 17, 21, 25 and 29).

### 3.2. Program workflow

The program MAELAS has been designed to read and write files for the Vienna Ab initio Simulation Package (VASP) code [34–36]. The workflow of MAELAS can be split into 5 steps: (i) cell relaxation, (ii) test of MAE, (iii) generation of distorted cells and spin directions, (iv) calculation of the energy with VASP, and (v) calculation of magnetostrictive coefficients and magnetoelastic constants. In Fig.2 we show a diagram with a summary of the MAELAS workflow. In the first step, it performs a full cell relaxation (ionic positions, cell volume, and cell shape) of the input unit cell. If one wants to use non-relaxed lattice parameters (like experimental ones), then this step can be skipped. In the next step, it is recommended to check if it is possible to obtain a realistic value of MAE for the ground state (not distorted cell). To do so, MAELAS generates the VASP input files to calculate MAE for two spin directions given by the user which should be compared with available experimental data. In the third step, MAELAS performs a symmetry analysis with pymatgen library [37] to determine the crystal system, redefine the structure using the same IEEE lattice convention as in AELAS [32] and then generates a set of volume-conserving distorted cells and spin directions.
Table 2: Selected cell length (β) and magnetization directions (α₁, α₂) in MAELAS to calculate the anisotropic magnetostrictive coefficients according to Eq[30]. The first column shows the crystal system and the corresponding lattice convention set in MAELAS based on the IEEE format[32]. The second column presents the equation of the relative length change that we used in Eq[30] for each crystal system. In the last column we show the values of the parameter η that is defined in Eq[30]. The symbols a, b, c correspond to the lattice parameters of the relaxed (not distorted) unit cell.

| Crystal system | Eq | Magnetostriiction coefficient | β | α₁ | α₂ | η |
|----------------|----|--------------------------------|----|----|----|----|
| Cubic (I)      |     | λ₀₀₁                           | (0, 0, 1) | (0, 0, 1) | (1, 0, 0) | ½ |
| a∥[k, b]∥[y, c]∥[z] |     | λ₁₁₁                           | (1, 0, 0) | (1, 0, 0) | (1, 0, 0) | ½ |
| Hexagonal (I)  | Eq[16] | λ₁₁₂                           | (1, 0, 0) | (1, 0, 0) | (1, 0, 0) | ½ |
| a∥[k, c]∥[z]   |     | λ₂₂₂                           | (0, 0, 1) | (0, 0, 1) | (0, 1, 0) | 1 |
| b = (−½, √6, 0) |     | λ₃₃₃                           | (1, 0, 0) | (0, 1, 0) | (0, 1, 0) | 1 |
| Trigonal (I)   | Eq[20] | λ₁₁₂                           | (1, 0, 0) | (0, 0, 1) | (0, 0, 1) | 1 |
| a∥[k, c]∥[z]   |     | λ₂₂₂                           | (0, 0, 1) | (0, 0, 1) | (0, 0, 1) | 1 |
| b = (−½, √6, 0) |     | λ₃₃₃                           | (1, 0, 0) | (0, 0, 1) | (0, 0, 1) | 1 |
| Tetragonal (I) | Eq[24] | λ₁₁₂                           | (1, 0, 0) | (1, 0, 0) | (1, 0, 0) | 1 |
| a∥[k, b]∥[y, c]∥[z] |     | λ₂₂₂                           | (0, 0, 1) | (0, 0, 1) | (0, 0, 1) | 1 |
| a = b ≠ c      |     | λ₃₃₃                           | (1, 0, 0) | (1, 0, 0) | (1, 0, 0) | 1 |
| Orthorhombic   | Eq[27] | λ₁₁₁                           | (1, 0, 0) | (0, 0, 1) | (0, 0, 1) | 1 |
| a∥[k, b]∥[y, c]∥[z] |     | λ₂₂₂                           | (0, 0, 1) | (0, 0, 1) | (0, 0, 1) | 1 |
| c < a < b      |     | λ₃₃₃                           | (1, 0, 0) | (0, 0, 1) | (0, 0, 1) | 1 |
|                 |     | λ₄₄₄                           | (0, 0, 1) | (0, 0, 1) | (0, 0, 1) | 1 |
|                 |     | λ₅₅₅                           | (1, 0, 0) | (0, 0, 1) | (0, 0, 1) | 1 |
|                 |     | λ₆₆₆                           | (0, 0, 1) | (0, 0, 1) | (0, 0, 1) | 1 |

The symbols a, b, c correspond to the lattice parameters of the relaxed (not distorted) unit cell.
according to Table 2. The crystal symmetry can also be set manually. In the fourth step, one should run the VASP calculations using these inputs. In order to help in this task, MAELAS generates some bash scripts to run all calculations with VASP automatically. Finally, MAELAS analyzes the calculated energies and fits them to a quadratic function (see Section 3.1) in order to obtain the magnetostrictive coefficients. If the elastic tensor is provided in the format given by the program AELAS [32], then the magnetoelastic constants are also calculated from the relations given in Section 2. Detecting possible calculation failures on fly is a very important feature of an automated high-throughput code. For instance, MAELAS prints a warning message when the R-squared of the quadratic curve fitting is lower than 0.98. It also automatically generates figures showing the quadratic curve fitting and the energy difference between states with spin directions $\alpha_2$ and $\alpha_1$ versus the cell length along $\beta$, so that the users can check the results easily.

Figure 2: Workflow of the program MAELAS and its connection with the program AELAS [32].
3.3. Computational details

The MAELAS code is written in Python3, and its source and documentation files are available in GitHub repository [38]. The DFT calculations are performed with VASP code, which is an implementation of the projector augmented wave (PAW) method [39]. We use the interaction potentials generated for the Perdew-Burke-Ernzerhof (PBE) version [40] of the Generalized Gradient Approximation (GGA). We follow the recommended procedure to determine MAE with spin-orbit coupling included non-self-consistently. Namely, a first collinear spin-polarized job (without spin-orbit coupling) is performed to calculate the wavefunction and charge density, and then a second non-collinear spin-polarized job is performed in a non-self-consistent manner, by switching on the spin-orbit coupling, reading the wavefunction and charge density generated in the collinear job, and defining the spin orientation through the quantisation axis (SAXIS-tag) [41]. The number of bands included in the non-collinear job should be twice the number of bands in the collinear job. By default, the code sets the tetrahedron method with Blöchl corrections for smearing in the calculation of MAE. The energy convergence criterion of the electronic self-consistency was chosen as $10^{-9}$ eV/cell, while the force convergence criterion of ionic relaxation was used, with all forces acting on atoms being lower than $10^{-3}$ eV/Å. MAELAS also generates the input file with the set of k-points in the reciprocal space for VASP by using an automatic Monkhorst–Pack k-mesh [42] gamma-centered grid with length parameter $R_k$ given by the user in the command line. Note the default settings generated by MAELAS for VASP might not work well for some materials, so that the user should check and tune them accordingly. For instance, in Section 4 we show some specific VASP settings and tests for few known materials. It is possible to use MAELAS with other DFT codes instead of VASP, after file conversion to VASP format files. Although, this process might require some extra work for the user.

4. Examples

In this section, we present some examples of the calculation of anisotropic magnetostrictive coefficients, elastic and magnetoelastic constants using MAELAS combined with AELAS [32] for a set of well-known magnetic materials. AELAS determines second-order elastic constants from the quadratic coefficients of the polynomial fitting of the energies versus strain relationships efficiently. For each material, we split the analysis into two parts. Firstly, we perform a cell relaxation, evaluate MAE and compute magnetostriction with MAELAS. In the second stage, we calculate the elastic constants with AELAS, and we use them as inputs.
to compute the magnetoelastic constants with MAELAS. To do so, we follow the workflow discussed in Section 3.2 (see Fig. 2). The results are shown in Tables 3, 4 and 5. All calculations correspond to zero-temperature.

Table 3: Anisotropic magnetostrictive coefficients and MAE calculated using the program MAELAS and measured in experiment (\(T \approx 0\) K) for a set of magnetic materials. In parenthesis we show the magnetostrictive coefficients with Mason’s definitions obtained using the relations given by Eq. A.2

| Material | Crystal system | DFT method | Magnetostriction coefficient | MAELAS (× 10^{-6}) | Exp. (× 10^{-6}) | MAE (\(\mu eV/atom\)) | MAELAS (\(\mu eV/atom\)) | Exp. (\(\mu eV/atom\)) |
|----------|----------------|-------------|-------------------------------|---------------------|-----------------|----------------------|----------------------|---------------------|
| BCC Fe   | Cubic (I)      | GGA         | \(\lambda_{001}\)             | 23                  | 26\(^{c}\)      | E(110) – E(001)     | 0.1                  | 1.0\(^{d}\)         |
|          |                |             | \(\lambda_{111}\)            | 19                  | -30\(^{c}\)     | E(111) – E(001)     | 0.21                 | 1.3\(^{d}\)         |
| HCP Co   | Hexagonal (I)  | SCAN        | \(\lambda_{111}^{\alpha1}\) (\(\lambda_{001}\)) | -85 (-78)           | 95 (-66\(^{e}\)) | E(100) – E(001)     | 53                   | 61\(^{b}\)         |
|          |                |             | \(\lambda_{111}^{\alpha2}\) (\(\lambda_{001}\)) | -115 (-92)          | -126 (-123\(^{e}\)) |                       |                     |                     |
|          |                |             | \(\lambda_{221}\) (\(\lambda_{c}\)) | 15 (115)            | 57 (126\(^{e}\)) |                       |                     |                     |
|          |                |             | \(\lambda_{222}\) (\(\lambda_{a}\)) | -19 (-1)            | -286 (-128\(^{e}\)) |                       |                     |                     |
| YCo      | Hexagonal (I)  | LSDA+U      | \(\lambda_{11}^{\alpha1},\lambda_{22}^{\alpha1}\) | -90                 | 115 (-92)       | E(100) – E(001)     | 365                  | 567\(^{e}\)         |
|          |                |             | \(\lambda_{22}^{\alpha2}\) | 76                  | 141             |                       |                     |                     |
| Fe\(_2\)Si | Trigonal (I)  | GGA         | \(\lambda_{11}^{\alpha1},\lambda_{22}^{\alpha1}\) | -9                  | 15              | E(100) – E(001)     | -38                  |                     |
|          |                |             | \(\lambda_{12}^{\alpha1}\) | 8                   | 28              |                       |                     |                     |
| L1\(_0\)FePd | Tetragonal (I) | GGA        | \(\lambda_{11}^{\alpha1},\lambda_{22}^{\alpha1}\) | -21                 | 79              | E(100) – E(001)     | 106                  | 181\(^{f}\)         |
|          |                |             | \(\lambda_{12}^{\alpha1}\) | 31                  | 28              |                       |                     |                     |
|          |                |             | \(\lambda_{12}^{\alpha2}\) | 106                 | 106             |                       |                     |                     |
| YCo      | Orthorhombic   | LSDA+U      | \(\lambda_{11},\lambda_{22},\lambda_{33}\) | -11                 | 32              | E(100) – E(001)     | 22                   | -23                 |
|          |                |             | \(\lambda_{22},\lambda_{33}\) | 70                  | 70              | E(010) – E(001)     |                     |                     |
|          |                |             | \(\lambda_{33},\lambda_{44}\) | -74                 | -74             |                       |                     |                     |
|          |                |             | \(\lambda_{44},\lambda_{55}\) | -30                 | -30             |                       |                     |                     |
|          |                |             | \(\lambda_{55},\lambda_{66}\) | 7                   | 7               |                       |                     |                     |
|          |                |             | \(\lambda_{66},\lambda_{77}\) | 36                  | 36              |                       |                     |                     |
|          |                |             | \(\lambda_{77},\lambda_{88}\) | -20                 | -20             |                       |                     |                     |
|          |                |             | \(\lambda_{88},\lambda_{99}\) | 35                  | 35              |                       |                     |                     |

\(^{a}\)Ref.[43], \(^{b}\)Ref.[44], \(^{c}\)Ref.[45], \(^{d}\)Ref.[4], \(^{e}\)Ref.[46], \(^{f}\)Ref.[47], \(^{g}\)Ref.[48]
Table 4: Elastic and magnetoelastic constants calculated using the interface between AELAS and MAELAS codes. The third column shows the DFT method used to compute the elastic constants. The experimental elastic constants of BCC Fe and HCP Co were measured at $T \approx 0$ K and $T \approx 300$ K, respectively. The experimental magnetoelastic constants were estimated using the experimental elastic constants (seventh column) and the experimental magnetostrictive coefficients in Table 3 via the relations given in Section 2. The sixth column presents calculations of the elastic constants available in the Materials Project database [49, 50].

| Material | Crystal system | DFT method | Elastic constant | AELAS (GPa) | Mat.Proj. (GPa) | Expt. (GPa) | Magnetoelastic constant | MAELAS (MPa) | Expt. (MPa) |
|----------|----------------|-------------|------------------|-------------|----------------|-------------|-----------------------|---------------|-------------|
| BCC Fe   | Cubic (I)      | GGA         | $c_{11}$         | 286         | 247$^a$        | 243$^b$     | $b_1$                 | -4.6          | -4.1        |
|          |                |             | $c_{12}$         | 152         | 150$^a$        | 138$^b$     | $b_2$                 | -5.9          | 10.9        |
|          |                |             | $c_{44}$         | 103         | 97$^a$         | 122$^b$     |                       |               |             |
| HCP Co   | Hexagonal (I)  | GGA         | $c_{11}$         | 459         | 358$^a$        | 307$^d$     | $b_{21}$              | -39.1         | -31.9       |
|          |                |             | $c_{12}$         | 179         | 165$^a$        | 165$^d$     | $b_{22}$              | 33.6          | 25.5        |
|          |                |             | $c_{13}$         | 132         | 114$^a$        | 103$^d$     | $b_3$                 | -4.2          | -8.1        |
|          |                |             | $c_{33}$         | 487         | 409$^a$        | 358$^d$     | $b_4$                 | 4.5           | 42.9        |
|          |                | SCAN        | $c_{11}$         | 118         | 95$^a$         | 75$^d$      |                       |               |             |
|          |                |             | $c_{12}$         | 212         | 189            |             |                       |               |             |
|          |                |             | $c_{13}$         | 633         | 113$^c$        |             |                       |               |             |
|          |                |             | $c_{44}$         | 239         | 48$^c$         |             |                       |               |             |
| YCo$_5$  | Hexagonal (I)  | GGA         | $c_{11}$         | 208         | 192$^a$        |             | $b_{21}$              | 14.9          |             |
|          |                |             | $c_{12}$         | 103         | 123$^c$        |             | $b_{22}$              | -10.4         |             |
|          |                |             | $c_{13}$         | 114         | 113$^c$        |             | $b_3$                 | -8.0          |             |
|          |                |             | $c_{33}$         | 270         | 262$^c$        |             | $b_4$                 | -13.6         |             |
|          |                | LSDA+U      | $c_{11}$         | -63         |                 |             | $b_{21}$              | 13.9          |             |
|          |                |             | $c_{12}$         | 363         |                 |             | $b_{22}$              | -7.9          |             |
|          |                |             | $c_{13}$         | 115         |                 |             | $b_3$                 | 32.5          |             |
|          |                |             | $c_{33}$         | 249         |                 |             | $b_4$                 | -12.4         |             |

$^a$Ref. [51], $^b$Ref. [52], $^c$Ref. [53], $^d$Ref. [54], $^e$Ref. [55]
Table 5: Elastic and magnetoelastic constants calculated using the interface between AELAS and MAELAS codes. The third column shows the DFT method used to compute the elastic constants. The experimental elastic constants of L1₀ FePd were measured at \( T \approx 300 \) K. The sixth column presents calculations of the elastic constants available in the Materials Project database [49, 50].

| Material      | Crystal system | DFT method | Elastic constant | AELAS (GPa) | Mat.Proj. (GPa) | Expt. (GPa) | Magnetoelastic constant | MAELAS (MPa) | Expt. (MPa) |
|---------------|----------------|------------|------------------|-------------|----------------|-------------|-------------------------|--------------|-------------|
| Fe₂Si         | Trigonal (I)   | GGA        | \( c_{11} \)     | 428         | 415\(^a\)      | 404         | \( b_{11} \)              | 3.1          |             |
|               |                |            | \( c_{12} \)     | 164         | 169\(^a\)      | 160         | \( b_{12} \)              | -4.2         |             |
|               |                |            | \( c_{13} \)     | 133         | 133\(^a\)      | 130         | \( b_{3} \)               | -0.7         |             |
|               |                |            | \( c_{14} \)     | -27         | -25\(^a\)      | 26          | \( b_{4} \)               | 3.3          |             |
|               |                |            | \( c_{33} \)     | 434         | 428\(^a\)      | 420         | \( b_{14} \)              | -1.4         |             |
|               |                |            | \( c_{44} \)     | 118         | 107\(^a\)      | 105         | \( b_{14} \)              | -0.4         |             |
| L₁₀ FePd      | Tetragonal (I) | GGA        | \( c_{11} \)     | 324         | 293\(^b\)      | 284         | \( b_{11} \)              | -2.4         |             |
|               |                |            | \( c_{12} \)     | 67          | 62\(^b\)       | 61          | \( b_{12} \)              | -15.2        |             |
|               |                |            | \( c_{33} \)     | 264         | 254\(^b\)      | 243         | \( b_{13} \)              | -7.9         |             |
|               |                |            | \( c_{44} \)     | 101         | 99\(^b\)       | 98          | \( b_{4} \)               | -5.6         |             |
| YCo           | Orthorhombic   | GGA        | \( c_{11} \)     | 76          | 94\(^d\)       | 93          | \( b_{1} \)               | -0.9         |             |
|               |                |            | \( c_{12} \)     | 45          | 61\(^d\)       | 60          | \( b_{2} \)               | 0.6          |             |
|               |                |            | \( c_{13} \)     | 48          | 44\(^d\)       | 43          | \( b_{3} \)               | -5.0         |             |
|               |                |            | \( c_{22} \)     | 102         | 93\(^d\)       | 92          | \( b_{4} \)               | 5.7          |             |
|               |                |            | \( c_{23} \)     | 55          | 56\(^d\)       | 55          | \( b_{5} \)               | 0.9          |             |
|               |                |            | \( c_{33} \)     | 141         | 121\(^d\)      | 120         | \( b_{6} \)               | 1.5          |             |
|               |                |            | \( c_{44} \)     | 40          | 38\(^d\)       | 37          | \( b_{7} \)               | -5.0         |             |
|               |                |            | \( c_{55} \)     | 27          | 29\(^d\)       | 28          | \( b_{8} \)               | 1.1          |             |
|               |                | LSDA+U     | \( c_{11} \)     | 101         |                 |             | \( b_{1} \)               | -1.7         |             |
|               |                |            | \( c_{12} \)     | 65          |                 |             | \( b_{2} \)               | 1.2          |             |
|               |                |            | \( c_{13} \)     | 58          |                 |             | \( b_{3} \)               | -3.8         |             |
|               |                |            | \( c_{22} \)     | 94          |                 |             | \( b_{4} \)               | 4.3          |             |
|               |                |            | \( c_{23} \)     | 70          |                 |             | \( b_{5} \)               | -0.1         |             |
|               |                |            | \( c_{33} \)     | 138         |                 |             | \( b_{6} \)               | 2.3          |             |
|               |                |            | \( c_{44} \)     | 42          |                 |             | \( b_{7} \)               | -4.4         |             |
|               |                |            | \( c_{55} \)     | 29          |                 |             | \( b_{8} \)               | 1.1          |             |
|               |                |            | \( c_{66} \)     | 35          |                 |             | \( b_{9} \)               | -8.7         |             |

\(^a\)Ref. [56], \(^b\)Ref. [57], \(^c\)Ref. [58], \(^d\)Ref. [59]

4.1. BCC Fe

In the first example, we consider BCC Fe, which is described by Eq.10 since it is a cubic (I) system.

4.1.1. Cell relaxation, MAE and magnetostrictive coefficients

Here, we need to use a very large k-point mesh due to the low MAE of this material (\( \sim \mu \text{eV/atom} \)). After analyzing the convergence of MAE, we set the length parameter \( R_k = 185 \) for the generation of the automatic k-point mesh. Thus, for
the conventional cubic cell of the BCC Fe at equilibrium volume, the calculations were performed with a $65 \times 65 \times 65$ k-mesh with 274625 k-points in the Brillouin zone. The interactions were described by a PAW potential with 14 valence electrons within the PBE approximation to the exchange-correlation, and the PW basis was generated for an energy cut-off of 439.857 eV (50% larger than the default value). The relaxed lattice parameter is $a = 2.8263$ Å. The calculated values of MAE are $E(110) - E(001) = 0.1\mu\text{eV/atom}$ and $E(111) - E(001) = 0.21\mu\text{eV/atom}$ which are below the experimental values $1\mu\text{eV/atom}$ and $1.3\mu\text{eV/atom}$, respectively [44]. Concerning the magnetostrictive coefficients, we obtained $\lambda_{001} = 23 \times 10^{-6}$ and $\lambda_{111} = 19 \times 10^{-6}$, while the experimental values at $T = 4.2\text{K}$ are $\lambda_{001} = 26 \times 10^{-6}$ and $\lambda_{111} = -30 \times 10^{-6}$ [43]. We see that $\lambda_{001}$ is quite close to the experimental result, while $\lambda_{111}$ is in good agreement with previous DFT calculations [15, 60] but it has the opposite sign as the experimental value. In Fig.3, we show the quadratic curve fit to the energy versus cell length along $\beta = \left(\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}\right)$ with $\alpha_1 = \left(\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}\right)$ to calculate $\lambda_{111}$, as well as the energy difference between states with spin directions $\alpha_2 = \left(\frac{1}{\sqrt{2}}, 0, -\frac{1}{\sqrt{2}}\right)$ and $\alpha_1 = \left(\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}\right)$ against the cell length along $\beta = \left(\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}\right)$. We observe that the sign of the calculated $\lambda_{111}$ ($>0$) is the same as the sign of the derivative of the energy difference between states with spin directions $\alpha_2$ and $\alpha_1$ with respect to the cell length along $\beta$ evaluated at $l = l_2$, as it is expected from Eq.35. However, as we mentioned above, the sign of $\lambda_{111}$ is negative in the experiment. This deviation might be related to the location of the Fermi level in a region of majority band t2g density of states [61].
4.1.2. Elastic and magnetoelastic constants

To compute the elastic constants we make use of AELAS code [32]. As inputs, we use the same relaxed cell and VASP settings as in the calculation of magnetostriction, but without spin-orbit coupling and lower number of k-points \( R_k = 60 \) (\( 21 \times 21 \times 21 \) for the not distorted cell). Once we have the elastic constants, we use them as inputs to derive the magnetoelastic constants with MAELAS. The results are shown in Table 4, where we also include calculations of elastic constants available in the Materials Project database [49, 50] and experimental data [52]. We observe that the value of \( c_{11} = 286 \) GPa obtained with AELAS is significantly higher than the one in the Materials Project \( c_{11} = 247 \) GPa and in the experiment \( c_{11} = 243 \) GPa. Regarding the magnetoelastic constants, we see that the value for \( b_1 = -4.6 \) MPa generated with MAELAS is very close to the estimated experimental value \( b_1 = -4.1 \) MPa. However, we obtain a negative sign for \( b_2 = -5.9 \) MPa, while in the experiment it is positive \( b_2 = 10.9 \) MPa. This deviation is due to the wrong sign of the calculated \( \lambda_{111} \) that we have mentioned above, see Eq.11.

4.2. HCP Co

As a first example of hexagonal (I) system, we consider HCP Co.
4.2.1. Cell relaxation, MAE and magnetostrictive coefficients

For this material, we set the length parameter $R_k = 160$ for the generation of the automatic k-point mesh, which for the relaxed (not distorted) cell, results in a $75 \times 75 \times 40$ k-point grid with 250000 points in the Brillouin zone. All calculations were done with an energy cut-off 406.563 eV (50\% larger than the default one), 15 electrons in the valence states, and the meta-GGA functional SCAN \[62\], with aspherical contributions to the PAW one-centre terms. The relaxed lattice parameters are $a = b = 2.4561$ Å and $c = 3.9821$ Å. The calculated MAE for the relaxed cell is $E(100) - E(001) = 53\mu eV/atom$ which is quite close to the experimental value $61\mu eV/atom$ \[44\]. As it is shown in Table 3, the calculated magnetostrictive coefficients are also close to the experimental ones, except for $\lambda^{\varepsilon,2}$. Similarly, converting them into Mason’s definitions via the relations given by Eq.A.2, we see that only $\lambda_D = -1 \times 10^{-6}$ is significantly deviated from the experiment ($-128 \times 10^{-6}$) \[45\]. In fact, the direct calculation of $\lambda_D$ using $\mathbf{B} = \left( \frac{1}{\sqrt{2}}, 0, \frac{1}{\sqrt{2}} \right)$, $\mathbf{\alpha}_1 = \left( \frac{1}{\sqrt{3}}, 0, \frac{1}{\sqrt{3}} \right)$ and $\mathbf{\alpha}_2 = (0, 0, 1)$ gives $\lambda_D = -9 \times 10^{-6}$, which is also far from the experimental value. Fig 4 shows the quadratic curve fit to the energy versus cell length along $\mathbf{\beta} = (1, 0, 0)$ with $\mathbf{\alpha}_1 = \left( \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}} \right)$ to calculate $\lambda^{\alpha_1,2}$.

Figure 4: Calculation of $\lambda^{\alpha_1,2}$ for HCP Co using MAELAS. (Left) Quadratic curve fit to the energy versus cell length along $\mathbf{\beta} = (1, 0, 0)$ with spin direction $\mathbf{\alpha}_1 = \left( \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}} \right)$. (Right) Energy difference between states with spin directions $\mathbf{\alpha}_2 = \left( \frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0 \right)$ and $\mathbf{\alpha}_1 = \left( \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}} \right)$ against the cell length along $\mathbf{\beta} = (1, 0, 0)$. 

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4.2.2. Elastic and magnetoelastic constants

For the calculation of the elastic constants we use the same relaxed cell and VASP settings as in the calculation of magnetostriction, but without spin-orbit coupling and lower number of k-points \(R_k = 60\) (28 \(\times\) 28 \(\times\) 15 for the not distorted cell). In addition to SCAN, we also run calculations with GGA. In Table 4, we see that GGA gives better results than SCAN for both the elastic and magnetoelastic constants. The magnetoelastic constants obtained with GGA are in quite good agreement with the estimated experimental values, except for \(b_4\) which is one order of magnitude lower than in the experiment due to the low \(\lambda^{\varepsilon,2}\) given by MAELAS, see Table 3.

4.3. YCo

The compound YCo5 is a hexagonal (I) system with prototype CaCu5 structure (space group 191).

4.3.1. Cell relaxation, MAE and magnetostrictive coefficients

In this case, we use the simplified (rotationally invariant) approach to the LSDA+U [63] with parameters \(U = 1.9\) eV and \(J = 0.8\) eV for Co, and \(U = J = 0\) eV for Y given in Ref.[46]. For the calculation of the relaxed cell, MAE and magnetostrictive coefficients we used an automatic k-point mesh with length parameter \(R_k = 100\) centered on the \(\Gamma\)-point (23 \(\times\) 23 \(\times\) 25 for the not distorted cell), 11 and 9 valence states for Y and Co, respectively, and energy cut-off 375 eV. The cell relaxation leads to lattice parameters \(a = b = 4.9253\) Å and \(c = 3.9269\) Å. The calculated MAE is \(E(100) - E(001) = 365\mu\text{eV/atom}\) which is lower than the experimental value 567\(\mu\text{eV/atom}\) [46]. Andreev measured the magnetostriction along the a axis, finding that the magnitude of \(|\lambda^{\alpha1,2}|\) and \(|\lambda^{\alpha2,2}|\) can not be greater than \(10^{-4}\) [4]. We obtained \(\lambda^{\alpha1,2} = -90 \times 10^{-6}\) and \(\lambda^{\alpha2,2} = 115 \times 10^{-6}\) which are quite close to the experimental upper limit. In Fig 5 we present the quadratic curve fit to the energy versus cell length along \(\vec{B} = (0, 0, 1)\) with \(\vec{a}_1 = (0, 0, 1)\) to calculate \(\lambda^{\alpha2,2}\).
4.3.2. Elastic and magnetoelastic constants

The calculation of the elastic constants is performed using the same relaxed cell and VASP settings as for magnetostriction, but without spin-orbit coupling and lower number of k-points $R_k = 60$ ($14 \times 14 \times 15$ for the not distorted cell). In addition to LSDA+U, we also run calculations with GGA. In Table 4, we observe that LSDA+U leads to an unstable phase ($c_{11} - c_{12} < 0$), while GGA gives better results.

4.4. Fe$_2$Si

To illustrate the application of MAELAS to trigonal (I) systems, we apply it to Fe$_2$Si (space group 164) [64].

4.4.1. Cell relaxation, MAE and magnetostrictive coefficients

For the calculation of the cell relaxation, MAE and magnetostrictive coefficients we used an automatic k-point mesh with length parameter $R_k = 80$ centered on the $\Gamma$-point ($24 \times 24 \times 17$ for the not distorted cell), 14 and 4 valence states for Fe and Si, respectively, and energy cut-off 520 eV with PAW method and GGA-PBE. The relaxed lattice parameters are $a = 3.9249$ Å and $c = 4.8311$ Å. The calculated MAE is $E(100) - E(001) = -38 \mu$eV/atom (easy plane). Sun et al. reported MAE values with the screened hybrid Heyd-Scuseria-Ernzerhof (HSE06) functional smaller than with PBE for 2D Fe$_2$Si [65][66]. Chi Pui Tang et al. calculated some electronic properties for bulk Fe$_2$Si finding that the densities of states
in the vicinity of the Fermi level is mainly contributed from the d-electrons of Fe [67]. In Table 5 we observe that the overall anisotropic magnetostriction given by MAELAS is rather small, which makes this material interesting for high-flux core applications because it can reduce hysteresis loss [68].

Figure 6: Calculation of $\lambda^{\alpha_2,\alpha_1}$ for Fe$_2$Si using MAELAS. (Left) Quadratic curve fit to the energy versus cell length along $\beta = (0,0,1)$ with spin direction $\alpha_1 = (0,0,1)$. (Right) Energy difference between states with spin directions $\alpha_2 = (1,0,0)$ and $\alpha_1 = (0,0,1)$ against the cell length along $\beta = (0,0,1)$.

4.4.2. Elastic and magnetoelastic constants

As inputs for AELAS, we use the same relaxed cell and VASP settings as in the calculation of magnetostriction, but without spin-orbit coupling and lower number of k-points $R_k = 60$ ($18 \times 18 \times 12$ for the not distorted cell). In Table 5 we see that AELAS gives similar elastic constants as in the Materials Project [56]. The derived magnetoelastic constants are small due the low magnetostrictive coefficients.

4.5. L1$_0$ FePd

As an example of tetragonal (I) system, we calculate the anisotropic magnetostrictive coefficients of L1$_0$ FePd (space group 123).

4.5.1. Cell relaxation, MAE and magnetostrictive coefficients

For the calculation of the cell relaxation, MAE and magnetostrictive coefficients we used an automatic k-point mesh with length parameter $R_k = 100$ centered on the $\Gamma$-point ($37 \times 37 \times 27$ for the not distorted cell), 8 and 10 valence states for Fe and Pd, respectively, and energy cut-off 375 eV with PAW method.
and GGA-PBE. The relaxed lattice parameters are \( a = 2.6973 \, \text{Å} \) and \( c = 3.7593 \, \text{Å} \). We obtained a MAE \( E(100) - E(001) = 106 \mu\text{eV/atom} \) which is lower than in the experiment \( 181 \mu\text{eV/atom} \) [47]. The values of the obtained anisotropic magnetostrictive coefficients are shown in Table 3. Shima et al. reported a relative length change equal to \( 100 \times 10^{-6} \) along a-axis under a magnetic field in the same direction \( \vec{\beta} = \vec{\alpha} = (1, 0, 0) \) [48]. According to Eq. 24, this measurement corresponds to \( \lambda_{\alpha1,0} - \frac{\lambda_{\alpha1,2}}{3} + \frac{\lambda_{\gamma2}}{2} \). For the anisotropic part of this quantity, we obtained \(-\frac{\lambda_{\alpha1,2}}{3} + \frac{\lambda_{\gamma2}}{2} = 22.5 \times 10^{-6}\). In Fig. 7 we show the quadratic curve fit to the energy versus cell length along \( \vec{\beta} = (1, 0, 0) \) with \( \vec{\alpha}_1 = (1, 0, 0) \) to calculate \( \lambda_{\gamma2} \).

![Figure 7: Calculation of \( \lambda_{\gamma2} \) for L1_0 FePd using MAELAS. (Left) Quadratic curve fit to the energy versus cell length along \( \vec{\beta} = (1, 0, 0) \) with spin direction \( \vec{\alpha}_1 = (1, 0, 0) \). (Right) Energy difference between states with spin directions \( \vec{\alpha}_2 = (0, 1, 0) \) and \( \vec{\alpha}_1 = (1, 0, 0) \) versus the cell length along \( \vec{\beta} = (1, 0, 0) \).](image)

4.5.2. Elastic and magnetoelastic constants

The calculation of the elastic constants with AELAS is performed using the same relaxed cell and VASP settings as for magnetostriction, but without spin-orbit coupling and lower number of k-points \( R_k = 60 \) \((22 \times 22 \times 16 \) for the not distorted cell). As we see in Table 5 we obtain similar results as in the Materials Project database [57].

4.6. YCo

For the case of orthorhombic systems, we study the compound YCo (space group 63) [69].
4.6.1. Cell relaxation, MAE and magnetostrictive coefficients

In this case, we use the simplified (rotationally invariant) approach to the LSDA+U \[63\] with parameters $U = 1.9$ eV and $J = 0.8$ eV for Co, and $U = J = 0$ eV for Y in the same way as in YCo$_5$ \[46\]. For the calculation of the relaxed cell, MAE and magnetostrictive coefficients we used an automatic k-point mesh with length parameter $R_k = 90$ centered on the $\Gamma$-point ($22 \times 9 \times 23$ for the not distorted cell), 11 and 9 valence states for Y and Co, respectively, and energy cut-off 375 eV. The cell relaxation leads to lattice parameters $a = 4.0686$ Å, $b = 10.3157$ Å and $c = 3.8957$ Å. As we see in Table 3, both MAE and magnetostriction are quite small for this material.

![Figure 8: Calculation of $\lambda_5$ for YCo using MAELAS. (Left) Quadratic curve fit to the energy versus cell length along $\beta = (0, 0, 1)$ with spin direction $\alpha_1 = (1, 0, 0)$. (Right) Energy difference between states with spin directions $\alpha_2 = (0, 0, 1)$ and $\alpha_1 = (1, 0, 0)$ against the cell length along $\beta = (0, 0, 1)$.](image)

4.6.2. Elastic and magnetoelastic constants

The calculation of the elastic constants is performed using the same relaxed cell and VASP settings as for magnetostriction, but without spin-orbit coupling and lower number of k-points $R_k = 60$ ($15 \times 6 \times 15$ for the not distorted cell). In addition to LSDA+U, we also run calculations with GGA. In Table 5, we observe that both LSDA+U and GGA lead to similar elastic and magnetoelastic constants.

5. Conclusions and future perspectives

In summary, the program MAELAS offers computational tools to tackle the complex phenomenon of magnetostriction by automated first-principles calculations. It could be used to discover and design novel magnetostrictive materials by
a high-throughput screening approach. In particular, materials with giant magnetostriction (beyond conventional cubic and hexagonal systems), isotropic or very low magnetostriction (like FeNi alloys) might be of technological importance.

The preliminary tests of the program show quite encouraging results, although there is still room for improvement. Presently, we are working on new features of MAELAS and online visualization tools. Alternative approaches based on the direct calculation of magnetoelastic constants through linear fittings of the magnetoelastic energy versus strain would be worth exploring in the future.

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Appendix A. Conversion between different definitions of magnetostrictive coefficients for hexagonal (I)

The magnetostrictive coefficients for hexagonal (I) shown in Eq.[16] were defined by Clark [10]. However, one can find other definitions like those given by Mason [9], Birss [26], and Callen and Callen [11]. In this appendix, we show the conversion formulas between these definitions and those provided by Clark [10] (Eq.[16]).

Appendix A.1. Mason’s form

Based on a general thermodynamic function with stresses and intensity of magnetization as the fundamental variables [70], Mason derived the following
form of the relative length change \[9\]

\[
\frac{\Delta l}{l_0} \bigg|_\beta = \lambda_{\alpha1,0}^{\text{Mason}} (\beta_x^2 + \beta_y^2) + \lambda_{\alpha2,0}^{\text{Mason}} \beta_z^2 + \lambda_A \left[ (\alpha_x \beta_x + \alpha_y \beta_y)^2 - (\alpha_x \beta_x + \alpha_y \beta_y) \alpha_z \beta_z \right] \\
+ \lambda_B \left[ (1 - \alpha_z^2)(1 - \beta_z^2) - (\alpha_x \beta_x + \alpha_y \beta_y)^2 \right] \\
+ \lambda_C \left[ (1 - \alpha_z^2) \beta_z^2 - (\alpha_x \beta_x + \alpha_y \beta_y) \alpha_z \beta_z \right] + \lambda_D (\alpha_x \beta_x + \alpha_y \beta_y) \alpha_z \beta_z.
\]

(A.1)

These magnetostrictive coefficients are related to those defined in Eq.16 as \[10\]

\[
\lambda_{\alpha1,0}^{\text{Mason}} = \lambda_{\alpha1,0} + \frac{2}{3} \lambda_{\alpha1,2} \\
\lambda_{\alpha2,0}^{\text{Mason}} = \lambda_{\alpha2,0} + \frac{2}{3} \lambda_{\alpha2,2} \\
\lambda_A = -\lambda_{\alpha1.2} + \frac{1}{2} \lambda_{\gamma2} \\
\lambda_B = -\lambda_{\alpha1.2} - \frac{1}{2} \lambda_{\gamma2} \\
\lambda_C = -\lambda_{\alpha2.2} \\
\lambda_D = \frac{1}{2} \lambda_{\epsilon,2} - \frac{1}{4} \lambda_{\alpha1.2} + \frac{1}{8} \lambda_{\gamma2} - \frac{1}{4} \lambda_{\alpha2.2}.
\]

(A.2)

Note in the original work of Mason \[9\] the terms that describes the volume magnetostriction were not included. Here we added these terms \((\lambda_{\alpha1.0}^{\text{Mason}}, \lambda_{\alpha2.0}^{\text{Mason}})\) in order to fully recover the Eq.16.

Appendix A.2. Birss’s form

In 1959, Birss derived an equivalent equation of relative length change in this form \[26\]

\[
\frac{\Delta l}{l_0} \bigg|_\beta = Q_0 + Q_1 \beta_z^2 + Q_2 (1 - \alpha_z^2) + Q_4 (1 - \alpha_z^2) \beta_z^2 + Q_6 (\alpha_x \beta_x + \alpha_y \beta_y) \alpha_z \beta_z \\
+ Q_8 (\alpha_x \beta_x + \alpha_y \beta_y)^2.
\]

(A.3)
These magnetostrictive coefficients are related to those defined in Eq. 16 as [10] 

\[ Q_0 = \lambda^{\alpha 1,0} + \frac{2}{3} \lambda^{\alpha 1,2} \]

\[ Q_1 = \lambda^{\alpha 2,0} + \frac{2}{3} \lambda^{\alpha 2,2} - \lambda^{\alpha 1,0} - \frac{2}{3} \lambda^{\alpha 1,2} \]

\[ Q_2 = -\lambda^{\alpha 1,2} - \frac{1}{2} \lambda^{\gamma 2} \]

\[ Q_4 = \lambda^{\alpha 1,2} + \frac{1}{2} \lambda^{\gamma 2} - \lambda^{\alpha 2,2} \]

\[ Q_6 = 2\lambda^{\epsilon,2} \]

\[ Q_8 = \lambda^{\gamma,2} \]

(A.4)

Appendix A.3. Callen and Callen’s form

Callen and Callen obtained other equivalent form of the equation of relative length change by including two-ion interactions into the theory of magnetostriction arising from single-ion crystal-field effects [11]. It reads

\[ \frac{\Delta l}{l_0} \bigg|_{\beta}^{\alpha} = \frac{1}{3} \lambda_{11}^\alpha + \frac{1}{2\sqrt{3}} \lambda_{12}^\alpha \left( \alpha_z^2 - \frac{1}{3} \right) + 2\lambda_{21}^\alpha \left( \beta_z^2 - \frac{1}{3} \right) + \sqrt{3} \lambda_{22}^\alpha \left( \alpha_z^2 - \frac{1}{3} \right) \left( \beta_z^2 - \frac{1}{3} \right) + \lambda^\gamma \left[ \frac{1}{2} (\alpha_x^2 - \alpha_y^2)(\beta_x^2 - \beta_y^2) + 2\alpha_x\alpha_y\beta_x\beta_y \right] + 2\lambda^\epsilon (\alpha_x\alpha_z\beta_x\beta_z + \alpha_y\alpha_z\beta_x\beta_y), \]

(A.5)

These magnetostrictive coefficients are related to those defined in Eq. 16 as [11]

\[ \lambda_{11}^\alpha = 2\lambda^{\alpha 1,0} + \lambda^{\alpha 2,0} + 2\lambda^{\alpha 1,2} + \lambda^{\alpha 2,2} \]

\[ \lambda_{12}^\alpha = \frac{4}{\sqrt{3}} \lambda^{\alpha 1,2} + \frac{2}{\sqrt{3}} \lambda^{\alpha 2,2} \]

\[ \lambda_{21}^\alpha = -\frac{1}{2} \lambda^{\alpha 1,0} + \frac{1}{2} \lambda^{\alpha 2,0} \]

\[ \lambda_{22}^\alpha = -\frac{1}{\sqrt{3}} \lambda^{\alpha 1,2} + \frac{1}{\sqrt{3}} \lambda^{\alpha 2,2} \]

\[ \lambda^\epsilon = \lambda^{\epsilon,2} \]

\[ \lambda^\gamma = \lambda^{\gamma,2} \]

(A.6)
Appendix B. Conversion between different definitions of magnetostrictive coefficients for tetragonal (I)

Cullen et al. [5] derived the equation of relative length change given by Eq.24 for tetragonal (I) system. An equivalent equation was obtained by Mason earlier that reads [9]

\[
\frac{\Delta l}{l_0} = \lambda_{Mason}^{\alpha 1.0} (\beta_x^2 + \beta_y^2) + \lambda_{Mason}^{\alpha 2.0} \beta_z^2 + \frac{1}{2} \lambda_1 [(\alpha_x \beta_x - \alpha_y \beta_y)^2 - (\alpha_x \beta_y + \alpha_y \beta_x)^2] + (1 - \beta_z^2)(1 - \alpha_z^2) - 2\alpha_z \beta_z (\alpha_x \beta_x + \alpha_y \beta_y)] + 4\lambda_2 \alpha_z \beta_z (\alpha_x \beta_x + \alpha_y \beta_y) + 4\lambda_3 \alpha_x \alpha_y \beta_z \beta_x + \lambda_4 [\beta_z^2 (1 - \alpha_z^2) - \alpha_z \beta_z (\alpha_x \beta_x + \alpha_y \beta_y)] + \frac{1}{2} \lambda_5 [(\alpha_x \beta_y - \alpha_y \beta_x)^2 - (\alpha_x \beta_x + \alpha_y \beta_y)^2 + (1 - \beta_z^2)(1 - \alpha_z^2)].
\]

(B.1)

These magnetostrictive coefficients are related to those defined by Eq.24 in the following way

\[
\lambda_{Mason}^{\alpha 1.0} = \lambda^{\alpha 1.0} + \frac{2}{3} \lambda^{\alpha 1.2}
\]

\[
\lambda_{Mason}^{\alpha 2.0} = \lambda^{\alpha 2.0} + \frac{2}{3} \lambda^{\alpha 2.2}
\]

\[
\lambda_1 = -\lambda^{\alpha 1.2} + \frac{1}{2} \lambda^{\gamma 2}
\]

\[
\lambda_2 = \frac{1}{2} \lambda^{\varepsilon 2} - \frac{1}{4} \lambda^{\alpha 2.2} - \frac{1}{4} \lambda^{\alpha 1.2} + \frac{1}{8} \lambda^{\gamma 2}
\]

\[
\lambda_3 = \frac{1}{2} \lambda^{\delta 2} - \lambda^{\alpha 1.2}
\]

\[
\lambda_4 = -\lambda^{\alpha 2.2}
\]

\[
\lambda_5 = -\lambda^{\alpha 1.2} - \frac{1}{2} \lambda^{\gamma 2}.
\]

(B.2)

Note in the original work of Mason [9] the terms that describes the volume magnetostriction were not included. Here we added these terms \((\lambda_{Mason}^{\alpha 1.0}, \lambda_{Mason}^{\alpha 2.0})\) in order to fully recover the Eq.24.

Appendix C. Generation of the distorted unit cells

In this appendix we present the explicit form of the matrix transformations to generate the volume-conserving distorted unit cells for the calculation of each
magnetostrictive coefficient. The distorted cells are generated by multiplying the lattice vectors of the relaxed cell \( \mathbf{a} = (a_x, a_y, a_z), \mathbf{b} = (b_x, b_y, b_z), \mathbf{c} = (c_x, c_y, c_z) \) to the strain tensor \( \varepsilon_{ij} \) via the pymatgen library \[37\]

\[
\begin{pmatrix}
  a'_x & a'_y & a'_z \\
  b'_x & b'_y & b'_z \\
  c'_x & c'_y & c'_z 
\end{pmatrix}
= \begin{pmatrix}
  a_x & a_y & a_z \\
  b_x & b_y & b_z \\
  c_x & c_y & c_z 
\end{pmatrix}
\cdot \begin{pmatrix}
  \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\
  \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\
  \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} 
\end{pmatrix}, \quad (C.1)
\]

where \( a'_i, b'_i \) and \( c'_i (i = x, y, z) \) are the components of the lattice vectors of the deformed cell. The applied strain tensor in terms of the engineering shear strain \( \gamma_{ij} (i \neq j) \) is written as

\[
\varepsilon = \begin{pmatrix}
  \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\
  \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\
  \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} 
\end{pmatrix} = \begin{pmatrix}
  \frac{\gamma_{xx}}{2} & \frac{\gamma_{xy}}{2} & \frac{\gamma_{xz}}{2} \\
  \frac{\gamma_{yx}}{2} & \frac{\gamma_{yy}}{2} & \frac{\gamma_{yz}}{2} \\
  \frac{\gamma_{zx}}{2} & \frac{\gamma_{zy}}{2} & \frac{\gamma_{zz}}{2} 
\end{pmatrix}, \quad (C.2)
\]

where \( \varepsilon_{ij} = \varepsilon_{ji} \) and \( \gamma_{ij} = \gamma_{ji} (i \neq j) \). Additionally, the determinant of the strain tensor must be equal to 1 in order to conserve the volume of the cell

\[
|\varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} |
|\varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} |
|\varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} | = 1. \quad (C.3)
\]

**Appendix C.1. Cubic (I) system**

For cubic (I) systems MAELAS generates two set of distorted cells with tetragonal deformations along \( \mathbf{\beta} = (0, 0, 1) \) and trigonal deformations along \( \mathbf{\beta} = (\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}) \) to calculate \( \lambda_{001} \) and \( \lambda_{111} \), respectively (see Table 2). The strain tensors of these two deformation modes are

\[
\mathbf{\varepsilon}_{\mathbf{\beta}=(0,0,1)} = \begin{pmatrix}
  \frac{1}{\sqrt{1+\varepsilon}} & 0 & 0 \\
  0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\
  0 & 0 & 1 + \varepsilon 
\end{pmatrix}, \quad \mathbf{\varepsilon}_{\mathbf{\beta}=(\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}})} = \zeta \begin{pmatrix}
  \frac{1}{\varepsilon} & \frac{\varepsilon}{2} & \frac{\varepsilon}{2} \\
  \frac{\varepsilon}{2} & \frac{1}{\varepsilon} & \frac{\varepsilon}{2} \\
  \frac{\varepsilon}{2} & \frac{\varepsilon}{2} & \frac{1}{\varepsilon} 
\end{pmatrix} \quad (C.4)
\]

where \( \zeta = \sqrt{4/(4 - 3\varepsilon^2 + \varepsilon^4)} \). The parameter \( \varepsilon \) controls the applied strain and its maximum value can be specified through the command line of the program MAELAS, as well as the total number of distorted cells. Note in the trigonal deformation the engineering shear strain is given by \( \gamma = \zeta \cdot \varepsilon \). Here at very low values of \( \varepsilon \) (\( |\varepsilon| \ll 1 \)) we have \( \zeta \approx 1 \), so that \( \varepsilon \) is approximately the engineering shear strain (\( \gamma \approx \varepsilon \)).
Appendix C.2. Hexagonal (I) system

In the case of hexagonal (I), MAELAS generates 4 sets of distorted cells using the following strain tensors

\[
\varepsilon_{\beta=(1,0,0)}^{\lambda_{21}} = \begin{pmatrix}
1 + \varepsilon & 0 & 0 \\
0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\
0 & 0 & \frac{1}{\sqrt{1+\varepsilon}}
\end{pmatrix},
\varepsilon_{\beta=(0,0,1)}^{\lambda_{21}} = \begin{pmatrix}
\frac{1}{\sqrt{1+\varepsilon}} & 0 & 0 \\
0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\
0 & 0 & 1 + \varepsilon
\end{pmatrix},
\varepsilon_{\beta=(1,0,0)}^{\lambda_{22}} = \begin{pmatrix}
1 + \varepsilon & 0 & 0 \\
0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\
0 & 0 & \frac{1}{\sqrt{1+\varepsilon}}
\end{pmatrix},
\varepsilon_{\beta=(a,c)}^{\lambda_{22}} = \begin{pmatrix}
1 & 0 & \varepsilon c \\
0 & 1 & 0 \\
\frac{\varepsilon a}{2c} & 0 & 1
\end{pmatrix}
\]

where \( \omega = \frac{3}{4\sqrt{4-(\varepsilon^2)}} \), \( a \) and \( c \) are the lattice parameters of the relaxed (not distorted) unit cell. The fractions \( c/a \) and \( a/c \) were introduced in the strain tensor elements \( \varepsilon_{xz} \) and \( \varepsilon_{zx} \), respectively, in order to generate distortions that meet the property \( \beta = \frac{a+c}{a+c} = \frac{d'+c'}{|d'|+|c'|} \), where \( a' \) and \( c' \) are the lattice vectors of the distorted unit cell, see Fig. C.9.

Figure C.9: Sketch of the deformation generated by the strain tensor given by Eq. C.5 to calculate \( \lambda_{21} \). The purple line represents the relaxed cell with lattice parameters \( (a,b,c) \), while the red line stands for the distorted cell with lattice parameters \( (a',b',c') \). This distortion meets the property \( \beta = \frac{a+c}{a+c} = \frac{d'+c'}{|d'|+|c'|} \).
Appendix C.3. Trigonal (I) system

In the case of trigonal (I), MAELAS generates 6 sets of distorted cells using the following strain tensors

\[
\varepsilon_{\beta = (1,0,0)}^{\lambda^{a1,2}} = \begin{pmatrix}
1 + \varepsilon & 0 & 0 \\
0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\
0 & 0 & \frac{1}{\sqrt{1+\varepsilon}}
\end{pmatrix},
\varepsilon_{\beta = (0,0,1)}^{\lambda^{a2,2}} = \begin{pmatrix}
\frac{1}{\sqrt{1+\varepsilon}} & 0 & 0 \\
0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\
0 & 0 & 1 + \varepsilon
\end{pmatrix}
\]

Appendix C.4. Tetragonal (I) system

In the case of tetragonal (I), MAELAS generates 5 sets of distorted cells using the following strain tensors

\[
\varepsilon_{\beta = (1,0,0)}^{\lambda^{a1,2}} = \begin{pmatrix}
1 + \varepsilon & 0 & 0 \\
0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\
0 & 0 & \frac{1}{\sqrt{1+\varepsilon}}
\end{pmatrix},
\varepsilon_{\beta = (0,0,1)}^{\lambda^{a2,2}} = \begin{pmatrix}
\frac{1}{\sqrt{1+\varepsilon}} & 0 & 0 \\
0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\
0 & 0 & 1 + \varepsilon
\end{pmatrix}
\]

\[
(C.6)
\]

\[
\varepsilon_{\beta = \frac{(a,0,c)}{\sqrt{a^2+c^2}}}^{\lambda^{Y1}} = \begin{pmatrix}
1 + \varepsilon & 0 & 0 \\
0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\
0 & 0 & \frac{1}{\sqrt{1+\varepsilon}}
\end{pmatrix}
\]

\[
\varepsilon_{\beta = \frac{(a,0,c)}{\sqrt{a^2+c^2}}}^{\lambda^{Y2}} = \begin{pmatrix}
1 + \varepsilon & 0 & 0 \\
0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\
0 & 0 & \frac{1}{\sqrt{1+\varepsilon}}
\end{pmatrix}
\]

\[
(C.7)
\]
Appendix C.5. Orthorhombic system

For orthorhombic crystals, MAELAS generates 9 sets of distorted cells using the following strain tensors

\[
\varepsilon_{\lambda_1}^{\beta=(1,0,0)} = \begin{pmatrix} 1 + \varepsilon & 0 & 0 \\ 0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\ 0 & 0 & \frac{1}{\sqrt{1+\varepsilon}} \end{pmatrix}, \quad \varepsilon_{\lambda_2}^{\beta=(1,0,0)} = \begin{pmatrix} 1 + \varepsilon & 0 & 0 \\ 0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\ 0 & 0 & \frac{1}{\sqrt{1+\varepsilon}} \end{pmatrix}
\]

\[
\varepsilon_{\lambda_3}^{\beta=(0,1,0)} = \begin{pmatrix} \frac{1}{\sqrt{1+\varepsilon}} & 0 & 0 \\ 0 & 1 + \varepsilon & 0 \\ 0 & 0 & \frac{1}{\sqrt{1+\varepsilon}} \end{pmatrix}, \quad \varepsilon_{\lambda_4}^{\beta=(0,1,0)} = \begin{pmatrix} \frac{1}{\sqrt{1+\varepsilon}} & 0 & 0 \\ 0 & 1 + \varepsilon & 0 \\ 0 & 0 & \frac{1}{\sqrt{1+\varepsilon}} \end{pmatrix}
\]

\[
\varepsilon_{\lambda_5}^{\beta=(0,0,1)} = \begin{pmatrix} \frac{1}{\sqrt{1+\varepsilon}} & 0 & 0 \\ 0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\ 0 & 0 & 1 + \varepsilon \end{pmatrix}, \quad \varepsilon_{\lambda_6}^{\beta=(0,0,1)} = \begin{pmatrix} \frac{1}{\sqrt{1+\varepsilon}} & 0 & 0 \\ 0 & \frac{1}{\sqrt{1+\varepsilon}} & 0 \\ 0 & 0 & 1 + \varepsilon \end{pmatrix}
\]

\[
\varepsilon_{\lambda_7}^{\beta=(a,b,0)} = \omega \begin{pmatrix} 1 & \frac{eb}{2a} & 0 \\ \frac{ea}{2b} & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad \varepsilon_{\lambda_8}^{\beta=(a,0,c)} = \omega \begin{pmatrix} 1 & 0 & \frac{ec}{2a} \\ 0 & 1 & 0 \\ \frac{ea}{2b} & 0 & 1 \end{pmatrix}
\]

\[
\varepsilon_{\lambda_9}^{\beta=(0,b,c)} = \omega \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & \frac{ec}{2b} \\ 0 & \frac{eb}{2c} & 1 \end{pmatrix}
\]

(C.8)

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