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Abstract. The magnetostriiction of Lu$_2$Fe$_{17}$ single crystal has been measured along the main $\bar{a}$, $\bar{b}$ and $\bar{c}$ hexagonal axes between 4 and 295 K by applying strong pulsed magnetic fields along the easy $\bar{a}$-axis. The irreducible strains, $\alpha\varepsilon^{\alpha\beta}$ (volume) and $\alpha\varepsilon^{\alpha\beta}$ (c/a-ratio), were obtained. The corresponding magnetoelastic parameters and elastic constants were calculated through FLAPW approach on DFT. These calculations are analysed at the same time with the experimental results to obtain information about the origin of the two-ion magnetoelastic contributions.

1. Introduction

Despite the advances in modern electronic structure theory, magnetostriiction (MS), $\lambda$, has rarely attracted the attention due to its intrinsic complexity. In principle, the MS can be determined from the subtraction of two separate total energy optimizations, with magnetization oriented along two different crystallographic directions by using FLAPW approach on DFT [1]. It is known that the main difficulty of this procedure is that the magneto crystalline anisotropy (MCA) energy due to spin-orbit coupling (SOC) is very small in most systems. Under this background, we propose a magnetoelastic (MEL) study of the volume and c/a-ratio contributions to the MS for the hexagonal Lu$_2$Fe$_{17}$ single crystal.

2. Experimental details.

Lu$_2$Fe$_{17}$ single crystal was grown by Czochralsky technique and X-ray characterized. MS measurements were performed in strong magnetic fields of up to 31 T, in the range between 4 K and room temperature [2]. These measurements were obtained by using the strain gauge (SG) technique with an AC bridge [3]. The magnetoresistance of SG is rather substantial at so intense magnetic fields and low temperatures [4], and extreme care was exercised in order to compensate it. The MS modes, $\lambda(\bar{a},\bar{b})$, were measured along $\bar{b} = \bar{a}, \bar{b}$ and $\bar{c}$ crystallographic directions, keeping magnetization oriented along the easy axis, $\bar{a} = \bar{a}$ by using a magnetic field (see Figure 1 (a-c)). The MS modes were measured within the hexagonal basal-plane (see Fig. 1(a) and Fig. 1 (b)). They are close in value but slightly different from those obtained along the $\bar{c}$-axis, which are higher (see Fig. 1(c)).

3. Computational methods.

The hexagonal Lu$_2$Fe$_{17}$ has a P63/mmc as space group and $a = 8.3879$ Å and $c = 8.2870$ Å as lattice constants [5]. A well ordered (i.e., like a Th$_2$Ni$_{17}$-type) crystalline structure is assumed for the ab-initio calculations, although it is known the existence of disorder between Lu and Fe sublattices.
These calculations were done under LAPW approach on DFT, and the total energy is optimized allowing us to calculate the single-ion contributions to MS modes and the elastic constants.

3.1. LAPW approach.

The electronic structure code WIEN2k uses the FLAPW on DFT [6], where core and valence states are calculated self-consistently. The core states are treated as fully relativistic, for the spherical part of the potential, where the full-potential is used for valence states. The Kohn-Sham equations are solved self-consistently until the input and output of the charge density and total energy are less than $10^{-4}-10^{-5}$ e/a.u.³ and $10^{-5}$ Ry. The LDA method [7] and the generalized gradient approximation (GGA) of Perdew et al [8] are used for the exchange-correlation potential. The core states are treated fully relativistic by solving the Dirac equation, while the valence states are treated semi-relativistically [9]. Local orbital extensions [10], with covered basis of approximately 844 basis functions, are used to reduce linearization errors in Lu and Fe spheres. The self-consistent calculations were performed with 10000 k-points in the irreducible Brillouin-zone and the muffin-tin radii, $R_{MT}$, are 3.1 and 2.21 Å, for Lu and Fe spheres, respectively. The basis set is determined by a plane-wave cut-off energy of $R_{MT} K_{max} = 9.0$, which gives a good convergence. To investigate MS, the SOC is invoked as second-variational scheme by taking all states below the cut-off energy of -4.0 Ry.

3.2. Energy optimization.

3.2.1. Elastic constants.

Let be the total energy of the crystal ions (without SOC) as,

$$E_{tot}^{(0)} - \left( \frac{\partial E_{tot}}{\partial V} \right)_V (V-V_0) + E_{el},$$

where $E_{tot}^{(0)}$ is the total initial energy, $V_0$ the initial volume, $V$ the volume of strained lattice, $E_{el}$ the elastic energy and $\left( \frac{\partial E_{tot}}{\partial V} \right)_V$ the pressure on the crystal lattice under a volume deformation. To first order, the strained lattice is related with unstrained one by, $\bar{r} = (\bar{I} + \bar{\varepsilon}) \cdot \bar{r}_0$, where $\bar{I}$ is the identity matrix and $\bar{\varepsilon}$ is the strain tensor. According to Hooke's law, the elastic energy can be expressed up to second order strain components, $E_{el} = \bar{\varepsilon}^T \cdot \bar{C} \cdot \bar{\varepsilon}$, where $\bar{\varepsilon}$ and $\bar{C}$ are the cartesian strains and the elastic constants, written in Voigt two-suffix notation, respectively. Assuming that $(V-V_0)$-term in Eq.(1) is linear with respect to the strains, $(V-V_0) = V_0 \text{Tr}(\bar{\varepsilon})$, it is possible to obtain the elastic constants from Eq. (1) by calculating, $C_{ij} = (\partial E_{tot}/\partial \varepsilon_i \varepsilon_j)|_{\bar{V}_0}$. The elastic energy can also be written by using the irreducible representations, $\Gamma$, of the group, $G$, of the crystal, $E_{el} = \bar{\varepsilon}^T \cdot \bar{C}^{\Gamma} \cdot \bar{\varepsilon}^{\Gamma}/2$, where now $\varepsilon^{\Gamma}$ and $C^{\Gamma}$ are the irreducible strains and elastic constants [12]. The volume and $c/a$-ratio irreducible elastic constants, $C_{11}^{\Gamma}$ and $C_{44}^{\Gamma}$ were calculated under FLAPW approach on DFT by doing the optimization of the total energy under two symmetry irreducible deformations of the crystalline cell, $\varepsilon^{a,b}$ and $\varepsilon^{a,c}$, respectively.
3.2.2. Magnetostriction.

The magnetic interactions existing in a solid are related with the distance between the crystal ions. The coupling with the distance is called as magnetoelastic coupling (MELC), and so, it is the origin of the MS. Two kinds of MELC contributions can be considered: (a) single-ion interactions which are propagated by SOC, making MS strongly anisotropic; and b) two-ions interactions as exchange and dipolar can give rise relevant contributions to the volume and c/a-ratio MS modes [11,12].

The theoretical description of the MELC in the solids assumes that the elastic and magnetoelastic (MEL) energy are invariant under crystal group $\mathcal{G}$ of the solids [11]. So, the Eq. (1) can be rewritten under the irreducible representations of $\mathcal{G}$ as,

$$ E_{\text{tot}} = E_{\text{tot}}^{(0)} - \left( \hat{M}^T \cdot \hat{e}^r + \left( \hat{e}^r \right)^T \cdot \hat{C}^e \cdot \hat{e}^r \right) / 2 , $$

(2)

where a $\hat{z}$-axis is assumed as quantization axis, and $\hat{M}^T$ are the 0 K irreducible MEL parameters [11,12]. In this work, we only are interested in the volume and c/a-ratio MEL modes at 0 K, $M^\alpha_{11}$ and $M^\alpha_{22}$, so they can be written under the Callen&Callen’s theory as a sum of single-ion ($M$-modes) and two-ion (D-modes) contributions, i.e., $M^\alpha_{11} = D^\alpha_{11} + M^\alpha_{11}^0$ and $M^\alpha_{22} = D^\alpha_{22} + M^\alpha_{22}^0$ [11,12].

The equilibrium strains, $\hat{\epsilon}^r$, can be obtained under the strain minimization of Eq. (2). These strains can be written for any $\hat{a}$ orientation of the quantization axis (as for example, the magnetization orientation), $\hat{\epsilon}^r (\hat{a}) = \hat{R}(\hat{z} \rightarrow \hat{a}) \cdot \hat{\epsilon}^r$, being $\hat{R}(\hat{z} \rightarrow \hat{a})$ the transformation from $\hat{z}$-axis to $\hat{a}$-quantization axis [11]. Finally, the MS mode, measured along a $\beta$-direction, can be obtained as function of $\hat{\epsilon}^r (\hat{a})$ for an hexagonal system as [11,12],

$$ \lambda(\hat{a}, \hat{b}) = (1/3) \hat{\epsilon}^{\alpha\alpha_1}(\hat{a}) + \sqrt{3} (\beta^2 \epsilon_{22}^\alpha - 1/3) \hat{\epsilon}^{\alpha\alpha_2}(\hat{a}) + (1/2) (\beta_{x}^\alpha - \beta_{y}^\alpha) \hat{\epsilon}^{\alpha\beta}(\hat{a}) + \text{ shear terms.} $$

(3)

The single-ion volume and c/a-ratio irreducible MEL parameters, $M^\alpha_{11}$ and $M^\alpha_{22}$, can be calculated under FLAPW approach on DFT including SOC, by doing an energy optimization under $\epsilon^{\alpha\alpha_1}$ and $\epsilon^{\alpha\alpha_2}$ deformations of the crystalline cell. The torque method was used to minimize the remaining uncertainties resulting from SOC interaction [1].

4. Analysis

The Fig. 2(a) shows the temperature dependence of irreducible strains: $\hat{\epsilon}^{\alpha\alpha_1}(\hat{a})$, $\hat{\epsilon}^{\alpha\alpha_2}(\hat{a})$ and $\hat{\epsilon}^{\alpha\beta}(\hat{a})$, at the maximum applied field, with $\hat{a} = (1,0,0)$. These irreducible modes were obtained from: $\lambda(\hat{a}, \hat{a})$, $\lambda(\hat{a}, \hat{b})$ and $\lambda(\hat{a}, \hat{c})$ by using Eq. (3). The $\hat{\epsilon}^{\alpha\alpha_1}(\hat{a})$ and $\hat{\epsilon}^{\alpha\alpha_2}(\hat{a})$ modes have a remarkable feature around 250 K, very close to the antiferromagnetic ordering temperature [5], and decreasing both above 250 K, and being $\hat{\epsilon}^{\alpha\alpha_2}(\hat{a})$ practically null at room temperature. The 0 K values for these MS modes were obtained from temperature extrapolation, so, $\hat{\epsilon}^{\alpha\alpha_1}(\hat{a}) \approx 200 \mu\varepsilon$, $\hat{\epsilon}^{\alpha\alpha_2}(\hat{a}) \approx 270 \mu\varepsilon$ and $\hat{\epsilon}^{\alpha\beta}(\hat{a}) \approx 0 \mu\varepsilon$.

The Fig. 2(b) shows the energy optimization (without SOC) obtained from FLAPW approach calculations on DFT. The fit of $E_{\text{tot}}$ for $i=1$ with the third-order Birch-Murnaghan [13] equation state (see Fig. 2(b)), allow as to get the 0 K bulk modulus, which is about 140 GPa. Moreover, this optimization also allows us to calculate the corresponding irreducible elastic constants, just fitting $E_{\text{tot}}$ with a second order polynomial function, so, $C_{11} \approx 100$ GPa and $C_{22} \approx 145$ GPa. These values are in reasonable agreement with those obtained from Resonant Ultrasonic Spectroscopy [14], and the first one is relatively close to the bulk modulus previously calculated.

The Fig. 2(c) shows the energy optimization obtained from FLAPW approach calculations on DFT by using the SOC with magnetization aligned along [001] and [100] directions. Fig. 2(c) also shows the corresponding strain dependence of MCA, MCA= ($E_{\text{tot}}([100]) - E_{\text{tot}}([a,0,c])$)$_{a=0,c=1}$ (indeed, if we apply the Feyman-Hellman theorem, MCA can be evaluated from $(dE_{\text{tot}}/dt_e)_{t_e=0}$, with $0 \leq t_e \leq \pi/2$ [11]). So, the MEL parameters obtained are 110 MPa and -30 MPa for the volume and c/a-ratio energy optimization done, respectively. Assuming that the MCA used to calculate the MELC parameters has a single-ion origin, we can write that $M^\alpha_{11} \approx 110$ MPa and $M^\alpha_{22} \approx -30$ MPa. As a result of that, the two-ion MEL contributions could be estimated by using these MELC parameters and the 0 K values obtained from experimental results (see Ref. [15]), so, $D^\alpha_{11} \approx 160$ MPa and $D^\alpha_{22} \approx -10$ MPa.
Figure 2. (a) Temperature dependence of the hexagonal irreducible MS modes, \( \varepsilon^{a,i}(\vec{a}) \) (○), \( \varepsilon^{a,2}(\vec{a}) \) (●) and \( \varepsilon^{b,c}(\vec{b},\vec{c}) \) (♦), at the maximum applied field (30 T), as they are obtained from the MS modes, \( \lambda(\vec{a},\vec{b}) \) and \( \lambda(\vec{a},\vec{c}) \) by using Eq. (3); (b) \( E_{tot} \), without SOC, for the volume, \( \varepsilon^{a,1} \) (□), and the c/a-ratio, \( \varepsilon^{a,2} \) (◇), MS modes. The continuous line is the total energy fit with Birch-Murnaghan equation for \( i=1 \). The dash-dot lines are the quadratic polynomial fits of the \( E_{tot} \) with respect to irreducible MS modes \( \varepsilon^{a,1} \) and \( \varepsilon^{a,2} \); (c) \( E_{tot} \), optimized by using SOC for the volume MS mode, \( \varepsilon^{a,1} \) for \( \vec{a} = [001] \) (×) and \( \vec{a} = [100] \) (●) and for the c/a-ratio MS mode, \( \varepsilon^{a,2} \) for \( \vec{a} = [001] \) (■) and \( \vec{a} = [100] \) (●). The MCA is represented against the volume \( \varepsilon^{a,1} \) (▲) and the c/a-ratio, \( \varepsilon^{a,2} \) (▼), MS modes.

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5. References
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[15] \( \varepsilon^{a,1}(\alpha) = (M_{11}^{a,i} C_{11}^{a,i} - M_{12}^{a,i} C_{12}^{a,i}) < Y_2^g > + (D_0^{a,i} C_{22}^{a,i} D_0^{a,i} C_{22}^{a,i}) < S^2 > Y_2^g(\alpha) / (C_{11}^{a,i} C_{12}^{a,i} C_{12}^{a,i}) \) and \( \varepsilon^{a,2} = (M_{11}^{a,i} C_{11}^{a,i} - M_{12}^{a,i} C_{12}^{a,i}) < Y_2^g > + (D_0^{a,i} C_{22}^{a,i} D_0^{a,i} C_{22}^{a,i}) < S^2 > Y_2^g(\alpha) / (C_{11}^{a,i} C_{12}^{a,i} C_{12}^{a,i}) \), being \( < Y_2^g > \approx m^2, < S^2 > \approx m^2, m \) is reduced magnetization.