Effect of uniaxial pressure on helimagnetic structure in Lu$_2$Fe$_{17}$

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Abstract. The incommensurate helimagnetic structure (IHS) in the Lu$_2$Fe$_{17}$ intermetallics has been induced at 5 K by hydrostatic pressure above 0.3 GPa and characterized by a propagation vector $(00t_z)$. The observed dependence of $t_z$ on external variables is discussed within a simple model based on a complex dependence of exchange interactions $J_{n,m}$ $(n, m = f, j, k$ and $g)$ on distances between Fe-atoms in inequivalent positions $4f$, $12j$, $12k$ and $6g$ of the hexagonal $P6_3/mmc$ crystal structure. To verify this model, we have studied magnetization and representative neutron reflections of Lu$_2$Fe$_{17}$ single crystals under high hydrostatic pressure and uniaxial compression with stress $\sigma \parallel a$-axis and $\sigma \parallel c$-axis. Both, the metamagnetic transitions and magnetic satellites of the $(00t_z)$ type that verify the presence of IHS in Lu$_2$Fe$_{17}$ at 5 K were observed under hydrostatic pressure and under uniaxial stress $\sigma \parallel c$-axis only. We show that the dominant role in an appearance of IHS in the Lu$_2$Fe$_{17}$ under compression is played by the $J_{ff}$ and $J_{fj}$ exchange interactions.

1. Introduction
The family of the R$_2$Fe$_{17}$ ($R = \text{rare earth and Y}$) intermetallics is studied for decades, due to very diverse ferro- and ferrimagnetic properties that exhibit almost all members of this family [1,2]. The origin of a variety of magnetic structures, magnetoelastic effects and magnetocrystalline anisotropy is mostly ascribed to the rare earth element characteristics and their interaction with the ferromagnetic Fe-sublattice [2]. From this point of view, the R$_2$Fe$_{17}$ intermetallics with nonmagnetic R-elements ($R = \text{Lu and Y}$) offer a possibility to study magnetic behavior of the only Fe-sublattice in dependence on different external (T, P, H) variables.

The ground state of the Lu$_2$Fe$_{17}$ intermetallic compound is ferromagnetic, but, an incommensurate helimagnetic structure (IHS) was detected in a very narrow temperature range just below the Néel temperature $T_N$ [3]. A possible existence of non-collinear arrangement of magnetic Fe-moments in Fe-rich metallic systems (including Invar alloys) is discussed by theoreticians and searched for by experimentalists for almost a century. A similarity of magnetic behavior of Lu$_2$Fe$_{17}$ and Y$_2$Fe$_{17}$ intermetallics and Invar alloys is documented by their negative linear thermal expansions at temperatures below $T_N$ or the Curie temperature $T_C$, respectively. Both, the thermal expansion and the linear compressibility are strongly anisotropic with a pronounced decrease of the crystal $c$-parameter with increasing both, temperature and pressure [4]. Moreover, despite of a complex structure of these intermetallics, the ab-initio calculated local electronic structure of inequivalent Fe-sites exhibit identical features as the electronic structure of Invar alloys [5]. These exciting problems were the...
reason for intensive studies of both, the Lu$_2$Fe$_{17}$ and the Y$_2$Fe$_{17}$ intermetallics under pressure during the last decade.

The ferromagnetic ground state is totally suppressed in Lu$_2$Fe$_{17}$ and Y$_2$Fe$_{17}$ intermetallics by hydrostatic pressures above 0.3 GPa and 0.8 GPa, respectively [6-8]. The pressure induced IHS that is described by a propagation vector (00$\ell$) [3,7], is however, closely related to the original ferromagnetic structure and underlining hexagonal crystal structure of these intermetallics that is shown in figure 1. The ferromagnetic arrangement remains preserved in the basal ab-planes (easy magnetization plane), but, magnetization of the planes rotates as one moves along the crystal c-axis (that is the hard magnetization direction). Under the hydrostatic pressure, the ferromagnetic state in the intermetallics can be restored by external magnetic field only. The strong pressure dependence of the critical field $H_C$ applied in the basal plane of Lu$_2$Fe$_{17}$ along a-axis is shown in inset of figure 2, where magnetization isotherms measured at 5 K under different hydrostatic pressures are presented. Magnetic phase diagram of the Lu$_2$Fe$_{17}$ [9], constructed from magnetization measurements under hydrostatic pressure, is shown in figure 2.

A use of uniaxial stresses to a study of the Lu$_2$Fe$_{17}$ intermetallics in this work has been motivated by a need to understand on a more detailed level the role of direct exchange interactions in the occurrence of the non-collinear Fe-moments arrangement in these intermetallics. Based on the recent experimental data [10,11], we present a simple phenomenological model for the description of interplay of the Fe-Fe exchange interactions in Lu$_2$Fe$_{17}$. The observed effects of uniaxial stresses and hydrostatic pressures on the magnetic transitions in the Lu$_2$Fe$_{17}$ intermetallics are presented and discussed.

2. Experimental
The hexagonal Lu$_2$Fe$_{17}$ single crystal (P$6_3$/mmc space group, $a = 8.395$ Å, $c = 8.293$ Å) has been grown by the Czochralski method and oriented using the Laue X-ray diffraction method [12]. Miniature pressure cells (up to 1.2 GPa) were designed and fabricated with respect to the experimental arrangement of the E4 neutron diffractometer ($\lambda = 2.42$ Å) in connection with vertical and horizontal superconducting magnets installed at the Helmholtz Zentrum Berlin. A detail description of the hydrostatic CuBe cells for both, the microscopic (neutron scattering) and the macroscopic (magnetization) studies of single crystals under pressure can be found elsewhere [13]. In contrast to the isotropic effect of hydrostatic pressure, the uniaxial compression of a single crystal induces a
decrease of one crystal parameter (along the stress) and an increase of other (perpendicular) crystal parameters. The uniaxial cell presented in figure 3 allows to compress the Lu$_2$Fe$_{17}$ single crystal along the c-axis inside the E4 diffractometer (or a SQUID magnetometer) while keeping its easy magnetization direction (EMD) a-axis || H || axis of rotation of diffractometer (or magnetometer). To induce similar decrease of the crystal a-parameter, a single crystal can be compressed along the a-axis with the EMD b-axis || H || axis of rotation of diffractometer (or magnetometer). Both experimental arrangements enable the study of the (00t$_z$) propagation vector by reciprocal space scans due to the permanent orientation of the c-axis within the diffraction plane. The uniaxial pressure cell for neutron diffraction experiments is made of non-magnetic TiAl$_6$V$_4$ alloy and sliding parts of the prism (part 4 in figure 3) are made of a zirkon ceramics. The prism ensures compression of a sample in direction perpendicular to the axial force induced by a barrel spring (part 3 in figure 3). In a working position, the prism is fixed inside a tube (part 6 in figure 3) and all parts are inserted into the cell. The axial force is increased by a compression of the barrel spring by supporting screws (parts 2 in figure 3). The uniaxial pressure can be increased up to the yield stress of a studied material.

![Figure 3. Uniaxial pressure cell for neutron diffraction studies: 1 - cell, 2 - supporting screws, 3 - barrel spring, 4 - prism, 5 - washer, 6 - fixing tube.](image)

3. **Results**

3.1. *Experimental results*

The magnetization measurements were accompanied by representative neutron diffraction studies under identical external (T, P, H) conditions. Figure 4 shows reciprocal space scans along [001] direction in the vicinity of the nuclear (002) reflection at ambient pressure. Magnetic satellites (002+t$_z$) were observed at temperatures below the Néel temperature, T$_N$ = 275 K, down to a critical temperature for the ferromagnetic state, T$_C$ = 150 K. Similar magnetic satellites were also found around (101), (300) and (004) nuclear reflections. Satellites of a (t$_x$, t$_y$, 0) type have never been detected. The propagation vector t$_z$ decreases with decreasing temperature and intensity of the nuclear reflections increases after transition of Lu$_2$Fe$_{17}$ into ferromagnetic state (see figure 4). IHS induced by hydrostatic pressure or by uniaxial compression of crystal along c-axis exhibits magnetic satellites of the same type. Figure 5 shows magnetic satellites (002+t$_z$) with a much larger value of t$_z$. These reflections are induced at 5 K by hydrostatic pressure of 0.9 GPa and are suppressed by external magnetic field of 3 T without a change of intensity of the nuclear reflections.

The temperature dependence of the propagation vector t$_z$ of the pressure induced IHS is presented in figure 6. Generally, the value of t$_z$ increases with increasing pressure or uniaxial compression.
However, a linear decrease of $t_z$ with decreasing temperature at ambient pressure changes its character under hydrostatic pressure. In the case of uniaxial compression of crystal along c-axis, $t_z$ decreases with increasing temperature up to $T_c$ and then increases along the ambient $t_z(T)$ curve. To some extent, a more complex character of the $t_z(T)$ curve measured under hydrostatic pressure can be ascribed to a decrease of hydrostatic pressure inside the Cube cell with decreasing temperature. This effect is caused by a difference in thermal expansions of the CuBe cell and the pressure transmitting medium.

Figure 4. Temperature dependence of magnetic $(002 \pm t_z)$ satellites of nuclear $(002)$ reflection at ambient pressure.

Figure 5. Magnetic satellites of the nuclear $(002)$ reflection induced by pressure 0.9 GPa at 5 K at different magnetic fields.

The magnetization isotherms that were measured at 5 K under different pressures are shown in figure 7. Identical magnetization isotherms have been measured at ambient pressure and under uniaxial compression of crystal along a-axis up to the yield stress of Lu$_2$Fe$_{17}$. Hence, we were not able to induce IHS in this case by the uniaxial compression and these two magnetization isotherms at 5 K reflect the ferromagnetic ground state at ambient pressure and under uniaxial $\sigma \parallel a$-axis. The metamagnetic transitions that verify the presence of IHS in Lu$_2$Fe$_{17}$ at 5 K were observed under hydrostatic pressure and under uniaxial stress $\sigma \parallel c$-axis only, as shown in figure 7.

Figure 6. Temperature and pressure dependence of the propagation vector $t_z$.

Figure 7. Magnetization isotherms of Lu$_2$Fe$_{17}$ at 5K under different pressures.
3.2. Phenomenological model of the incommensurate helimagnetic structure

A simple phenomenological model of IHS in the Lu$_2$Fe$_{17}$ intermetallic compound is based on a complex dependence of exchange interactions $J_{n,m}$ ($n,m = f, j, k, g$) on distance $d_{\text{Fe-Fe}}$ between Fe atoms placed in the inequivalent 4f, 12j, 12k and 6g crystal positions, see figure 1. Values of $J_{n,m}$ were determined from experimental Mössbauer data recently [10,11]. These dependencies nicely follow the known Bethe-Slater curve with extremely sharp slope of negative exchange interaction below $d_{\text{Fe-Fe}} = 2.45\,\text{Å}$, see figure 8.

![Figure 8. Dependence of exchange interactions $J_{n,m}$ on interatomic distances between Fe$_n$ atoms placed in the inequivalent crystal lattice positions n= f, j, k, g](image)

| n   | $\mu_n$ | $J_{f,f}(0\,\text{GPa})$ | $J_{f,f}(1\,\text{GPa})$ | $J_{f,f}(\text{uniax})$ |
|-----|---------|--------------------------|--------------------------|------------------------|
| f   | 1.198   | -212 K                   | -121 K                   | -233 K                 | -190 K                 |
| g   | 1.039   | 45 K                     | 45 K                     | 50 K                   | 45 K                   |
| k   | 0.931   | 68 K                     | 68 K                     | 70 K                   | 68 K                   |
| j   | 0.965   | 10 K                     | 50 K                     | 53 K                   | 35 K                   |
| $-I_1/4I_2$ | 0.41 | 1.07                     | 0.59                     | 0.61                   |
| $t_z$ | 0.18 | --                       | 0.15                     | 0.14                   |
Considering the fact that the R-Fe interactions are much weaker than the direct exchange interaction $J_{n,m}$ between Fe atoms, we can assume that the experimentally determined dependence of $J_{n,m}$ on interatomic Fe-Fe distances presented in figure 8 is valid for all the $\text{R}_2\text{Fe}_{17}$ intermetallics. Then, the values of $J_{n,m}$ in Lu$_2$Fe$_{17}$ can be easily determined using the curve in figure 8 and known distances $d_{\text{Fe-Fe}}$ between Fe$_n$ atoms placed in the inequivalent positions, $n = f, g, k$ and $j$. The positions of Lu- and Fe-atoms in the studied Lu$_2$Fe$_{17}$ intermetallics were precisely determined by Rietveld method from neutron diffraction experiments at room temperature [14]. The received experimental values agree well with results in literature [3,15]. The refinement also gave relative values of Fe-moments in these inequivalent positions, $\mu_n$, with respect to $\mu_{\text{Fe}} = 2.15\mu_\text{B}/\text{at. Fe}$ [14,15]. Data are presented in table 1. Thermal expansion and compressibility of Lu$_2$Fe$_{17}$ were measured by both, the micro-strain gauges under hydrostatic pressures up to 1 GPa and by the neutron diffraction method [7,14]. The simple model of IHS is based on the layered crystal structure of Lu$_2$Fe$_{17}$, where the Lu+Fe$_{17}$ plane and the Fe$_0$+Fe$_8$ plane are the nearest neighbor planes of each the Fe$_f$ plane. The next nearest plane of the Fe$_f$ plane is again the Fe$_f$ plane, see figure 1. Due to a significant decrease of exchange interaction with increasing $d_{\text{Fe-Fe}}$ above 2.7 Å, the total exchange interaction between the planes is proportional to a sum of the exchange interactions between Fe$_f$ atom and its nearest neighbors, i.e. Fe$_f$, Fe$_g$, and Fe$_k$ atoms only. Hence, the exchange interactions $J_{fj}$, $J_{fg}$, $J_{fk}$ and $J_{fg}$ must be taken into account (black squares on the curve in figure 8). The positive values of the $J_{fj}$, $J_{fg}$ and $J_{fk}$ interactions ensure a collinear (ferromagnetic) arrangement of Fe-moments inside the Lu$^+$Fe$_f$ and the Fe$_0$+Fe$_8$ planes. IHS appears when a positive interaction between the Fe$_f$ plane and the nearest neighbor Lu$^+$Fe$_f$ and Fe$_0$+Fe$_8$ planes is overcome by a negative interaction between the next nearest neighbor Fe$_f$ planes. Then, Fe-moments in the Lu$^+$Fe$_f$ and Fe$_0$+Fe$_8$ planes are frustrated and magnetization of these planes rotates with respect to Fe-moments in the Fe$_f$ plane by an angle $\alpha$.

The exchange energy is proportional to the following sum:

$$E_{\text{ex}} \approx -\mu_{\text{Fe}}^{-2} \left( I_1 \cos(\alpha) + I_2 \cos(2\alpha) \right)$$

with $I_1 = \mu_n N_n \left( \mu_f N_{fj} + \mu_g N_{fg} + \mu_k N_{fk} \right)$ and $I_2 = \mu_n^2 N_n J_{ff}$, where $N_n$ is number of the relevant nearest neighbors and $\alpha$ is an angle between the orientation of Fe-moments in the adjacent Fe$_f$ planes. The minimum energy $E_{\text{ex}}$ with respect to the angle $\alpha$ is given by a general relation $\partial E_{\text{ex}}/\partial \alpha = 0$ that leads to a relation, $\cos(\alpha) = -I_1/4I_2$. Hence, IHS appears when the absolute value of the negative $I_2$ interaction is larger than one quarter of the positive interaction $I_1$. The results describing the appearance of IHS at temperatures 300 K and 5 K at ambient pressure and at temperature 5 K under hydrostatic pressure 1 GPa and uniaxial compression along c-axis ($\sigma = 0.3$ GPa) are collected in table 1. The distances $d_{\text{Fe-Fe}}$ were calculated from the refined crystal lattice parameters of Lu$_2$Fe$_{17}$, $a = 8.383$ Å, $c = 8.335$ Å at temperature 5 K and $a = 8.395$ Å, $c = 8.293$ Å at temperature 300 K. The used values of a linear compressibility, $\kappa_s = 3 \times 10^{-3}$ GPa$^{-1}$, $\kappa_c = 3.5 \times 10^{-3}$ GPa$^{-1}$ at 300K and $\kappa_s = 4.1 \times 10^{-3}$ GPa$^{-1}$, $\kappa_c = 7.5 \times 10^{-3}$ GPa$^{-1}$ at 5K, well agree with values of $\kappa_s$ and $\kappa_c$ at temperature 300 K presented in [15].

4. Discussion of the effects of hydrostatic and uniaxial pressures

The negative thermal expansion plays a very important role in the appearance of IHS in Lu$_2$Fe$_{17}$ at ambient pressure. Due to the larger $d_{ff}$ distance at temperature 5K than at 300 K, the negative $J_{ff}$ interaction at 5 K is not able to compete with the relatively strong positive interaction $J_{fj}$ and the ferromagnetic state is stable at 5 K at ambient pressure. Vice versa, a decrease of the lattice c-parameter with increasing temperature leads to a strengthening of the negative $J_{fj}$ interaction and a weakening of the positive $J_{fj}$ interaction, see table 1. As a result, IHS appears at temperatures above 150 K, up to $T_N$ at ambient pressure. In the case of Y$_2$Fe$_{17}$, the lattice parameters, $a = 8.466$ Å, $c = 8.315$ Å at room temperature, do not allow to reach a balance between $I_2$ and $I_1/4$. This intermetallic compound is ferromagnetic up to the temperature of its transition into paramagnetic state.
Under hydrostatic pressure, the high linear compressibility $\kappa_c$ at 5 K induces a very strong increase of the negative $J_{ff}$ interaction, see figure 8 that overcomes even an increase of the positive $J_{fj}$ interaction, see table 1. Hydrostatic pressure increases with increasing temperature in the clamped CuBe cell. This effect probably compensates a decrease of compressibility $\kappa_c$ with increasing temperature. So, the lattice parameters and the exchange interactions remain almost constant during heating of sample under hydrostatic pressure and values of the propagation vector $t_z$ in dependence on temperature exhibit a flat minimum only. The lower pressure, the more pronounced minimum has been observed as it was expected from the temperature dependence of $t_z$ at ambient pressure, see figure 6.

The effect of uniaxial compression of Lu$_2$Fe$_{17}$ single crystal along its c-axis on the negative $J_{ff}$ interaction at temperature 5 K is well comparable with the effect of hydrostatic pressure. However, a small increase of the crystal a-parameter induced by uniaxial compression leads to a smaller increase of the positive $J_{fj}$ interaction than in the case of hydrostatic pressure. The uniaxial compression seems to be more effective at 5 K than the compression by hydrostatic pressure. On the other hand, the uniaxial stress is not able to compensate the mentioned decrease of compressibility $\kappa_c$ with increasing temperature because the uniaxial stress $\sigma$ is not affected by increasing or decreasing temperature (there is no pressure transmitting medium). Hence, the effect of uniaxial compression weakens with increasing temperature up to a temperature where the effect of negative thermal expansion becomes dominant, see figure 6. Moreover, the maximum of uniaxial stress $\sigma$ is restricted by the yield stress of the studied material.

The direct exchange interaction exhibits the maximum in vicinity of $d_{4f}$ and $d_{6g}$ distances. Variations of these distances induced by pressure or temperature influence the relevant exchange interactions very slightly and so, $J_{4f}$ and $J_{6g}$ interactions do not play an important role in the studied effects. The same conclusion is also valid in the case of the positive $J_{jj}$ and $J_{kk}$ exchange interaction, see figure 8, and so, pressure does not destroy the ferromagnetic arrangement of Fe-moment inside the crystal Lu$^+$Fe and Fe$_k$+Fe$_g$ planes.

5. Conclusions
The ferromagnetic ground state of the Lu$_2$Fe$_{17}$ intermetallic compound has been suppressed by hydrostatic pressure and by uniaxial compression of the compound along its crystal c-axis. However, the ground state of Lu$_2$Fe$_{17}$ is not affected by a uniaxial compression along its crystal a-axis. The propagation vector (00$t_z$) of the pressure induced incommensurate helimagnetic structure exhibits the minimum as a function of temperature. This is a consequence of anisotropy of magneto-elastic properties of Lu$_2$Fe$_{17}$ that becomes evident in the pronounced temperature dependence of the linear compressibility $\kappa_c$ and in the negative thermal expansion along the crystal c-axis. The presented simple model of IHS elucidates almost all basic features of the effect of pressure on magnetic structures of the Lu$_2$Fe$_{17}$ intermetallics taking into account the direct exchange interaction between Fe atoms only. The model accentuates namely the dominant role of both, the negative exchange interaction $J_{ff}$ between Fe$_f$ atoms and the positive $J_{fj}$ exchange interaction in the appearance of IHS in the Lu$_2$Fe$_{17}$ intermetallic compound.

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