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Magnetization and neutron diffraction studies of Lu_2Fe_{17} under high pressure

J Kamarád[1], O Prokhnenko[1,3], K Prokeš[2] and Z Arnold[1]

[1] Institute of Physics ASCR, Na Slovance 2, 182 21 Prague 8, Czech Republic
[2] Hahn-Meitner-Institute, Glienickerstraße 100, D-141 09 Berlin, Germany

E-mail: prokhnen@fzu.cz

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Abstract

Combined magnetization and neutron diffraction measurements under high hydrostatic pressures up to 1 GPa were performed to study magnetic structures and their evolution in an Lu_2Fe_{17} single crystal. Pressures higher than 0.4 GPa suppress the collinear ferromagnetic ground state completely. The pressure induced magnetic structure can be described as a superposition of both incommensurate and commensurate magnetic components with propagation vectors $\tau_1 = (0; 0; \tau_{1z})$ and $\tau_2 = 0$. Mechanisms of the helimagnetic–ferromagnetic transitions induced by temperature and magnetic field in Lu_2Fe_{17} at ambient and high pressures are discussed.

1. Introduction

Recent studies of the Lu_2Fe_{17} compound with non-magnetic Lu atoms have brought appropriate and useful information on the magnetic behaviour of the Fe sublattice in natural multilayer R_2Fe_{17} intermetallic compounds (R is a rare earth element) [1–4]. The ground state of Lu_2Fe_{17} is ferromagnetic. However, Fe moments order helimagnetically above the transition temperature $\Theta_T = 140$ K. Above this temperature the magnetic Fe moments remain parallel within basal planes, but they change their direction from plane to plane going along the c-axis. The propagation vector $\tau_1 = (0; 0; \tau_{1z})$ is incommensurate with the crystal unit cell. This magnetic arrangement persists up to the Néel temperature $T_N = 275$ K. The helimagnetic phase manifests itself by the metamagnetic transition on magnetization isotherms at temperatures between Θ_T and T_N. Similar magnetic properties are typical for the Ce_2Fe_{17} compound [1, 5, 6]; however, only in the case of Lu_2Fe_{17} with non-magnetic Lu can one state that all effects originate from the Fe sublattice. This complex magnetic behaviour was attributed to the competition of positive and negative exchange interactions between the Fe ions on different crystallographic

[3] Address for correspondence: Institute of Physics ASCR, Cukrovarnicka 10, 162 53 Prague 6, Czech Republic.

sites [1]. Similar magnetic behaviour was observed in purely ferromagnetic Y_2Fe_{17} under high pressures only [7–9].

An extraordinary sensitivity of the magnetic structures in the R_2Fe_{17} compounds with R = Y, Ce, Lu to changes of the interatomic distances has been verified by magnetization and powder neutron diffraction studies under high hydrostatic pressures [3, 4, 6–10]. A very slight decrease of T_N under pressure ($dT_N/dP \sim -0.27$ K GPa^{-1}) is almost identical for all studied intermetallics. However, the ferromagnetic arrangement of Fe moments below Θ_T is suppressed relatively easily in all intermetallic compounds. Critical pressures for disappearance of the ferromagnetic structures are below 0.4 GPa in the case of Lu_2Fe_{17} and Ce_2Fe_{17} and below 1.2 GPa in the case of Y_2Fe_{17}. In contrast, an antiferromagnetic phase is induced by pressure and its range of stability expands down to the lowest temperatures. The pressure induced antiferromagnetic phase was detected by the metamagnetic transitions on magnetization curves and by magnetic superlattice reflections in neutron diffraction experiments. The magnetic phase diagram of Lu_2Fe_{17} based on magnetic measurements pointed to the presence of two different antiferromagnetic phases under high pressure [3], but they were not distinguished by the powder neutron diffraction measurements [4]. Moreover, the value of saturation magnetization at ambient pressure was not reached under pressure even at a field of 5 T.

In this work we present the combined microscopic (neutron diffraction) and macroscopic (magnetization) studies of the pressure–temperature–field evolution of the magnetic structures in a single crystal of Lu_2Fe_{17} under high hydrostatic pressure.

2. Experimental details

A hexagonal Lu_2Fe_{17} single crystal (a = 8.395 Å, c = 8.293 Å) has been grown by the Czochralski method. Details of the material preparation and its characterization can be found elsewhere [11]. A unique experimental set-up—neutron diffraction under pressures up to 1 GPa in magnetic fields up to 6.5 T—was used for the observation of the pressure induced magnetic structures and their changes with the magnetic field and with the temperature from 5 K up to 300 K. The single crystal of Lu_2Fe_{17} was fixed in a non-magnetic CuBe pressure cell in a position that ensured its orientation with the a-axis ∥ field H ∥ axis of rotation of the double-axis E4 diffractometer (λ = 2.44 Å) installed at HMI (Berlin). A mixture of mineral oils was used as a pressure-transmitting medium in the cell. The pressure values were determined at room temperature using a manganin pressure sensor. A decrease of pressure with decreasing temperature was monitored previously by changes of lattice constants of NaCl and it does not exceed 0.25 GPa. The diffractometer was equipped with a vertical superconducting magnet capable of generating 6.5 T.

Magnetization under pressure was measured using the same sample orientation as described above. The measurements were performed in a miniature CuBe hydrostatic pressure cell in a SQUID magnetometer. The pressure values were determined at low temperatures using the known pressure dependence of the critical temperature of the superconducting state of the Pb sensor placed inside the cell. The magnetization has been measured under pressures up to 1 GPa in magnetic fields up to 5 T at temperatures from 5 to 300 K. The changes of the pressure inside the pressure cells during temperature cycles have been taken into account.

3. Results and discussion

Neutron diffraction measurements on the single crystal of Lu_2Fe_{17} at ambient pressure confirmed the existence of three magnetic phases in Lu_2Fe_{17}: ferromagnetic, helimagnetic and

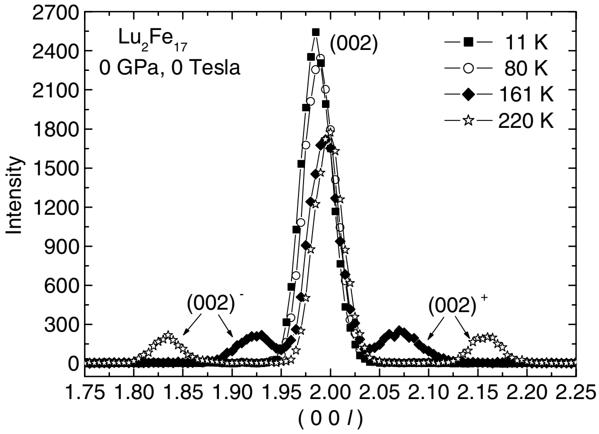

Figure 1. The 00l-scan at different temperatures at ambient pressure.

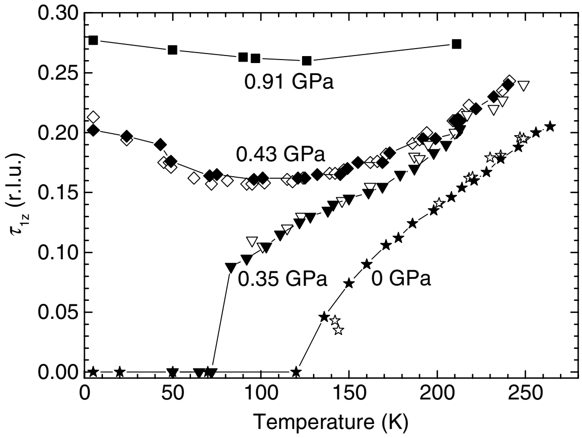

Figure 2. Temperature dependence of the z-component of the propagation vector τ_{1z} at different pressures. Solid symbols correspond to the values of τ_{1z} determined from the positions of $(002)^{\pm}$ and $(300)^{\pm}$ on cooling. Open symbols correspond to the τ_{1z} values on heating.

paramagnetic. A magnetic contribution is clearly observed in the ordered magnetic phases. Figure 1 shows l-scans through the reciprocal space (along the [00l]-direction) performed around $l = 2$ at various temperatures and ambient pressure. The ferromagnetic ground state is characterized by a significant magnetic contribution on top of nuclear reflections. The appearance of the superlattice reflections above 140 K points to the transition from the ferromagnetic to the helimagnetic phase (figure 1). The magnetic satellites have also been observed in the l-scans around (101), (300) and (004) nuclear reflections. This is the first time that all the magnetic satellites have been directly indexed as $(hkl) \pm \tau_1$ (marked as $(hkl)^{\pm}$ throughout the further text), where τ_1 is a propagation vector along the c-axis, $\tau_1 = (0; 0; \tau_{1z})$. The value of τ_{1z} is strongly temperature dependent and incommensurate with the crystal unit cell (figure 2). It decreases from $\tau_{1z} = 0.205$ r.l.u. at 265 K down to zero (in the ferromagnetic phase). In terms of the helimagnetic structure, the decrease of τ_{1z} down to zero with decreasing temperature indicates an increase of the helix length up to infinity,

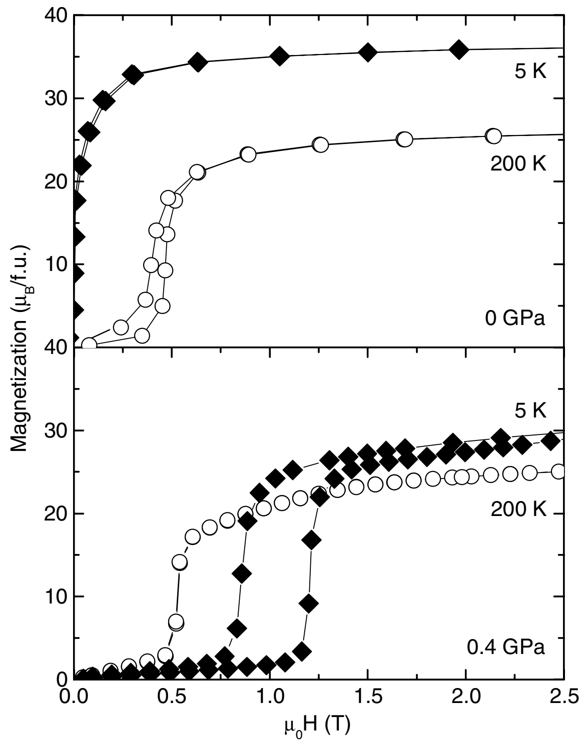

Figure 3. Magnetization isotherms at 5 and 200 K at ambient pressure and under pressure of 0.4 GPa.

and consequently a continuous transition into a collinear ferromagnetic structure below Θ_T at ambient pressure.

Application of a magnetic field along the *a*-axis of Lu_2Fe_{17} at 200 K results in the metamagnetic transition from the helimagnetic to the field induced ferromagnetic phase (figure 3). The critical field H_C of the metamagnetic transition increases with increasing temperature up to 0.5 T at 250 K. Microscopically, the metamagnetic transition was observed as a sudden drop of intensities of all the observed magnetic satellites down to zero at the magnetic fields above 0.6 T. However, a clearly detectable magnetic contribution to the nuclear reflection intensities was found only in the case of (101) reflection. Moreover, the value of τ_{1z} is practically independent of magnetic field in contrast to its strong temperature dependence. All these features show different character of the helimagnetic–ferromagnetic transitions induced by the temperature or the magnetic field at ambient pressure. The transformation of the magnetic structure due to the magnetic field goes without change of the spiral length. This can be realized, for example, through a distorted elliptical spiral. However, further investigations are necessary to solve this problem unambiguously.

The suppression of the ferromagnetic ground state in Lu_2Fe_{17} was observed under high hydrostatic pressure higher than 0.4 GPa in both magnetization and neutron diffraction measurements. Metamagnetic transitions on magnetization isotherms were detected at temperatures below T_N down to 5 K (figure 3). The metamagnetic behaviour of magnetization below and above 100 K is, however, different. Firstly, at pressures higher than 0.4 GPa, the value of the critical field H_C of the metamagnetic transition exhibits a minimum around 100 K.

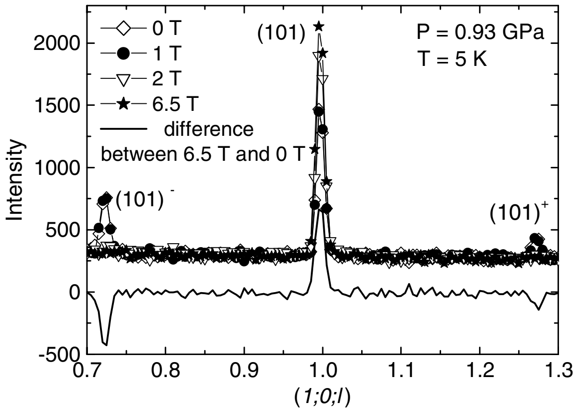

Figure 4. The 10*l*-scan at 5 K in different magnetic fields under pressure 0.93 GPa.

This seems to indicate an interphase between the low temperature pressure induced magnetic phase (LTP) and the high temperature one (HTP). Secondly, LTP exhibits a pronounced field hysteresis unlike the transitions in HTP (figure 3). Application of pressure leads to an increase of H_C in both phases.

Neutron diffraction measurements under pressure clarified the suppression of the ferromagnetic state in Lu_2Fe_{17}. At pressures above 0.4 GPa, the magnetic satellites $(101)^{\pm}$, $(300)^{\pm}$, $(002)^{\pm}$, $(004)^{\pm}$ corresponding to the propagation vector $\tau_1 = (0; 0; \tau_{1z})$ were observed in the whole temperature region from T_N down to 5 K. Figure 4 visualizes a reciprocal space scan along the *l*-direction performed at various fields around (101) at 5 K and maximum applied pressure of 0.93 GPa. Magnetic satellites $(101)^{\pm}$ are clearly observed on both sides of the nuclear peak. The propagation vector of the magnetic satellites is $\tau_{1z} = 0.277$.

Temperature and pressure evolution of the pressure induced magnetic structures is presented in figure 2 by a temperature dependence of τ_{1z} under pressure. Increase of the pressure leads to the increase of the propagation vector that is typical for all R_2Fe_{17} intermetallics with non-magnetic R-elements [4, 9, 10]. In terms of helical structure, this reflects the increase of the angle between Fe moments in the consecutive layers or the increase of negative exchange strength under pressure. Taking into account known negative thermal expansion along the *c*-axis in Lu_2Fe_{17} [12], both the pressure effect on τ_{1z} above Θ_T and the temperature effects on τ_{1z} at ambient pressure are understandable in terms of a simple Bethe–Slater model of the dependence of exchange interaction on Fe–Fe interatomic distances. At the lowest applied pressure of 0.35 GPa, a range of stability of the helimagnetic phase is expanded down to 80 K. Below this temperature Lu_2Fe_{17} undergoes a discontinuous transition into the ferromagnetic state as can be seen by a sudden drop of τ_{1z} down to zero at 80 K. Enhanced intensities of measured (101), (300), (002) and (004) nuclear reflections also confirm the creation of the ferromagnetic arrangement below 80 K under pressure of 0.35 GPa. The dependence of the propagation vector becomes complex under pressure above 0.4 GPa. τ_{1z} exhibits a wide minimum centred at 100 and 115 K at 0.4 GPa and 0.93 GPa, respectively. This minimum coincides with one in the temperature dependence of H_C under pressure, and similarly, it indicates an interphase between LTP and HTP. Both minima reflect a competition between positive and negative exchange interactions. However, direct thermal expansion and powder neutron diffraction measurements under high hydrostatic pressure showed monotonic changes of the lattice parameters in Lu_2Fe_{17} [12]. This means that the Bethe–Slater model alone cannot be used to explain the complex temperature behaviour of τ_{1z}

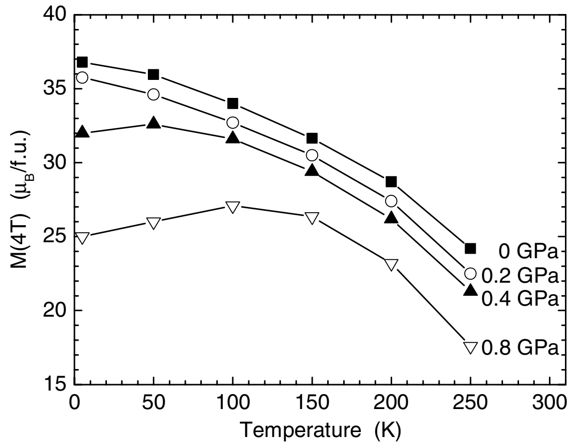

Figure 5. Temperature dependence of the magnetization at 4 T under pressure.

under pressure. Almost identical effects were also observed in Ce_2Fe_{17} and Y_2Fe_{17} compounds under hydrostatic pressure [9, 10].

Macroscopically, the decrease of saturated magnetization in Lu_2Fe_{17} at low temperatures under pressures above 0.4 GPa is much higher than that at high temperatures. The value of $\mathrm{d}\ln M/\mathrm{d}P$ at 5 K is the highest observed among Fe-rich intermetallic compounds [13]. The decrease of the magnetization with pressure above 0.4 GPa becomes smaller with increasing temperature and values of $\mathrm{d}\ln M/\mathrm{d}P$ become well comparable with the ones at 5 K and pressure lower than 0.4 GPa. Thus, the magnetization under pressures above 0.4 GPa has a tendency to be reduced below 100 K (figure 5). Such temperature behaviour of magnetization reflects in R–Fe intermetallics the presence of an antiferromagnetic sublattice at low temperatures. This strongly supports our two-phase concept involving LTP and HTP in Lu_2Fe_{17} under pressure.

Neutron diffraction measurements show that in addition to magnetic satellites there is a magnetic contribution on the top of (101), (002), (300) and (004) reflections at 5 K and 0.43 GPa in LTP, whereas the same reflections stay unchanged (paramagnetic) in HTP. This again clearly indicates the difference between LTP and HTP. Based on analysis of all the data, we can describe the HTP structure below T_N as the pure incommensurate helical arrangement of Fe moments with the propagation vector $\tau_1 = (0; 0; \tau_{1z})$ at both ambient and high pressure. The pressure induced LTP clearly exhibits the incommensurate helimagnetic component; however, a commensurate component described by $\tau_2 = 0$ seems to modify the magnetic structure of LTP in contrast to HTP.

Finally, the helimagnetic components in both phases respond similarly to the magnetic field. This is shown in figure 4 by a field dependence of the magnetic $(101)^{\pm}$ satellites under pressure. Though intensity of the $(101)^{\pm}$ satellites decreases with the field, their position does not change. Similarly to ambient pressure results, the value of τ_{1z} is independent of the applied magnetic field.

4. Conclusion

In conclusion, our combined neutron diffraction and magnetic studies of the Lu_2Fe_{17} single crystal under pressure showed that

(i) the ferromagnetic ground state is suppressed completely by pressure higher than 0.4 GPa;

(ii) the ferromagnetically ordered layers of iron in Lu_2Fe_{17} become ordered helimagnetically under pressure and two incommensurate helical structures are observed;
(iii) the high temperature phase (HTP) exhibits the incommensurate helimagnetic arrangement of the Fe moments in a limited temperature range below T_N at ambient and high hydrostatic pressures. Its propagation vector τ_1 decreases with decreasing temperature;
(iv) the low temperature phase (LTP) exhibits a remarkable field hysteresis of metamagnetic transitions from helimagnetic into field induced ferromagnetic structures. Its propagation vector τ_1 increases with decreasing temperature. The helimagnetic arrangement is modified in this phase by the commensurate component described by $\tau_2 = 0$.

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