Pressure study of magnetism in (Lu0.8Ce0.2)2Fe17 and Lu2Fe16.5Ru0.5 single crystals

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Abstract. The antiferromagnetic state observed in Lu2Fe17 at elevated temperatures is stabilized down to the ground state in (Lu0.8Ce0.2)2Fe17 and Lu2Fe16.5Ru0.5. The Néel temperature $T_N$ decreases from 275 K in Lu2Fe17 to 247 K and 208 K in (Lu0.8Ce0.2)2Fe17 and Lu2Fe16.5Ru0.5, respectively. Both compounds exhibit the easy-plane magnetic anisotropy and a metamagnetic transition at fields below 1 T. External pressure pushes the transition towards higher field and reduces the magnetization as well as $T_N$ to 234 K and 185 K for (Lu0.8Ce0.2)2Fe17 and Lu2Fe16.5Ru0.5, respectively, under pressure 0.8 GPa. The pressure and the substitution effects can be understood in terms of exchange interaction varying strongly with changes of the lattice parameter $c$.

1. Introduction

The very short Fe-Fe distances $d_{\text{FeFe}}$ between Fe atoms in selected inequivalent Fe positions, $d_{\text{FeFe}} \sim 239$ pm or 244 pm (shorter than $d_{\text{FeFe}} \sim 250$ pm in bcc Fe), are special features of the rare-earth intermetallic compounds $R_2Fe_17$. For this reason, a negative exchange interaction between the Fe atoms can coexist together with positive interactions in the case of larger $d_{\text{FeFe}}$ distances between Fe-atoms in basal planes. The total Fe-Fe exchange interaction is positive, however, the existence of these competitive interactions leads to unexpectedly low $T_C$ in compounds with so high content of Fe. The Lu2Fe17 compound with the smallest non-magnetic rare-earth element and hexagonal crystal structure of the Th2Ni17 type exhibits an incommensurate helimagnetic structure with the collinear Fe moments in basal planes below the Néel temperature $T_N = 274$ K [1,2]. The component $t_z$ of propagation vector $(00t_z)$ decreases continuously down to zero with decreasing temperature and a length of the magnetic helix increases to infinity at transition into ferromagnetic phase below 130 K. Simultaneously, the lattice parameter $c$ increases and a strength of the negative interactions decreases.

Such a delicate balance of ferro- (F) and antiferro- (AF) magnetic interactions is very sensitive to external conditions. Therefore, Lu3Fe17 is a very apt compound to study interplay of magnetic field, temperature, external pressure and substitution effects on magnetism of the high-Fe-content intermetallics. Just recently, the F state has been totally suppressed in Lu3Fe17 by external hydrostatic pressure [3,4]. Most of substitutions in the Fe sublattice (Al, Si, Cr, Co, Ni) and substitution of another non-magnetic atoms (Y) for Lu stabilize the F state and increase the magnetic ordering temperature.
On the other hand, external hydrostatic pressure stabilizes the AF state and reduces $T_N$ due to a strengthening of inter-plane AF and a weakening of the intra-plane F interactions by an induced decrease of the lattice $c$- and $a$-parameters.

In the present study we report on the rare cases where substitutions for Fe and Lu work together with pressure, i.e. they destabilize the F state and induce the AF structure in whole range of magnetic ordering.

2. Experimental

The single crystals of $(\text{Lu}_{0.8}\text{Ce}_{0.2})_2\text{Fe}_{17}$ and Lu$_2\text{Fe}_{16.5}\text{Ru}_{0.5}$ were prepared by the Czochralski method in a tetra-arc furnace in the same way as the parent compound Lu$_2\text{Fe}_{17}$ [2]. At ambient pressure, the magnetization isotherms were measured in SQUID and PPMS magnetometers (Quantum Design) along both principal axes of the hexagonal structure in fields up to 9 T at temperature range 5-300 K. Measurements under pressure were carried out in a miniature pressure piston-cylinder cell developed for a use in the SQUID magnetometer. The single crystals were oriented inside the cell with the $a$-axis (EMD) along magnetic field. The non-magnetic Cu-Be pressure cell with a mineral oil as a pressure-transmission medium allows us to apply hydrostatic pressure up to 1 GPa. Details of construction and operation of the cell, including a pressure determination using a pressure shift of the superconducting critical temperature of Pb, are described elsewhere [5]. All the magnetization curves were corrected for the demagnetization effect.

3. Results and discussion

Figure 1a) shows that the magnetization isotherms of $(\text{Lu}_{0.8}\text{Ce}_{0.2})_2\text{Fe}_{17}$ exhibit the metamagnetic transition started just below 1 T even at ambient pressure. The metamagnetic transition is clearly of the first order with a wide hysteresis. Magnetization reaches its saturation value 35 $\mu_B$ within approx. 1 T field interval above the transition. With increasing pressure, the transition field $H_c$ increases, while the magnetization gain across the transition $\Delta M$, the width of hysteresis $\Delta H_c$ and the Néel temperature $T_N$ (Fig. 1b) decrease. Qualitatively similar behavior, with larger drops of $T_N$ and saturation magnetization only, has been observed in Lu$_2\text{Fe}_{16.5}\text{Ru}_{0.5}$ (Fig. 2). Pressure dependencies of the magnetic characteristics of both compounds are presented in Fig. 3.

![Fig. 1: a) Magnetization of $(\text{Lu}_{0.8}\text{Ce}_{0.2})_2\text{Fe}_{17}$ vs. field along the $a$-axis at 5 K and b) Temperature dependence of magnetization of $(\text{Lu}_{0.8}\text{Ce}_{0.2})_2\text{Fe}_{17}$ in field 0.01 T under different pressures.](image-url)
be understood in terms of varying lattice parameter $c$. External pressure suppresses the F state in both Lu$_2$Fe$_{17}$ and Ce$_2$Fe$_{17}$. Despite of the larger unit-cell volume of Ce$_2$Fe$_{17}$ compared to Lu$_2$Fe$_{17}$ (due to larger parameter $a$), the F state is less stable in Ce$_2$Fe$_{17}$ where the $c$ parameter is smaller than in Lu$_2$Fe$_{17}$. Moreover, purely AF ground state was observed in Ce$_2$Fe$_{17}$ crystal [6] while this was not seen in Lu$_2$Fe$_{17}$. Lattice parameters of Lu$_2$Fe$_{17}$, (Lu$_{0.8}$Ce$_{0.2}$)$_2$Fe$_{17}$ and Ce$_2$Fe$_{17}$ are $a = 839.5$ pm, $c = 829.4$ pm; $a = 841.7$ pm, $c = 827.0$ pm; and $a = 848.9$ pm, $c = 827.2$ pm, respectively. (For Ce$_2$Fe$_{17}$ crystallizing in the rhombohedral Th$_2$Zn$_{17}$ structure the $c$ parameter 1241 pm is divided by factor of 1.5 to compare with hexagonal crystals). The $c$ parameter and consequently a distance between Fe atoms in “dumbbell” positions (along $c$ direction) are the same in (Lu$_{0.8}$Ce$_{0.2}$)$_2$Fe$_{17}$ and Ce$_2$Fe$_{17}$. That provides very similar magnetic properties (AF ground state with metamagnetic transition in 1 T [6]) despite of a considerable difference in the $a$ parameter. (Lu$_{0.8}$Ce$_{0.2}$)$_2$Fe$_{17}$ behaves as Lu$_2$Fe$_{17}$ uniaxially compressed by a “bias” chemical pressure (which is equivalent to approx. 0.4 GPa of hydrostatic pressure as follows from a comparison with pressure-study results on Lu$_2$Fe$_{17}$ [4]).

These speculations are not applicable to Lu$_2$Fe$_{16.5}$Ru$_{0.5}$ having the lattice parameters $a = 840.7$ pm, $c = 830.2$ pm so simply as in the case of (Lu$_{0.8}$Ce$_{0.2}$)$_2$Fe$_{17}$. A slight expansion along the $c$-axis (confirmed by $c = 831.2$ pm in compound with higher Ru content, Lu$_2$Fe$_{16}$Ru [7]) is observed upon the Ru substitution for Fe at room temperature. However, $T_N$ of this compound lies far below room temperature and thermal expansion of paramagnetic Lu$_2$Fe$_{16.5}$Ru$_{0.5}$ is positive in contrast with the anomalous negative thermal expansion of all R$_2$Fe$_{17}$ in the ordered states. Taking into account thermal expansion of Lu$_2$Fe$_{17}$ [8], the lattice parameters of Lu$_2$Fe$_{16.5}$Ru$_{0.5}$ and Lu$_2$Fe$_{17}$ are identical at the Néel temperatures. Since Ru atomic radius is larger than Fe one, the Ru atoms should not occupy the “dumbbells” positions and the AF interactions should not be weakened by the substitution. On the other hand, saturation magnetization Lu$_2$Fe$_{16.5}$Ru$_{0.5}$ is much lower than an estimated one assuming that Ru is a non-magnetic dilutor or a carrier of an antiparallel magnetic moment of a realistic magnitude. It is reasonable to conclude that this substitution modifies considerably the band structure of Lu$_2$Fe$_{17}$. This leads to decrease of the Fe magnetic moment, to a lower positive magnetostriction and to a less pronounced increase of the $c$ parameter with decreasing temperature. Therefore, the AF interactions remain strong and the F ground state cannot be reached in Lu$_2$Fe$_{16.5}$Ru$_{0.5}$ even at low temperatures.

Fig. 2: a) Magnetization of Lu$_2$Fe$_{16.5}$Ru$_{0.5}$ vs. field along the $a$-axis at 5 K and b) Temperature dependence of magnetization of Lu$_2$Fe$_{16.5}$Ru$_{0.5}$ in field 0.01 T under different pressures.
We can conclude that interplay between the dominant negative exchange interaction of Fe atoms in the "dumbbell" positions and the slight positive exchange interaction of other Fe atoms is an origin of the helimagnetism in the Lu₂Fe₁₇-based intermetallics. The observed pressure and substitution effects can be explained taking into account the anomalous and anisotropic thermal expansion induced by the anomalous and anisotropic magnetostriction. In agreement with this conclusions, pressure behavior of (Lu₀.₈Ce₀.₂)₂Fe₁₇ and Lu₂Fe₁₆.₅Ru₀.₅ compounds can be well compared with behavior of the parent Lu₂Fe₁₇ compound under external pressure [3,4].

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