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Magnetic properties of $Lu_2Fe_{16}X$ (X = Fe, Ni, Cr, Si) intermetallics under high hydrostatic pressure

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Abstract

We present very significant effects of a partial substitution of Ni, Cr and Si for Fe on magnetic and magnetoelastic properties of the mother Lu_2Fe_{17} intermetallics. The ferromagnetic state is stabilized by the substitution in all the $Lu_2Fe_{16}X$ (X = Ni, Cr, Si) intermetallics. Their Curie temperature T_C is by about 100 K higher than the Néel temperature T_N of Lu_2Fe_{17}. The magnetoelastic phenomena of the $Lu_2Fe_{16}X$ single crystals were studied under high hydrostatic pressure using a non-magnetic pressure CuBe cell in a SQUID magnetometer. The effect of pressure on T_C of the $Lu_2Fe_{16}X$ intermetallics coincides with changes of T_C that are induced by hydrogenation in $Lu_2Fe_{17}H_x$. Pressure induced changes of saturated magnetization M_S of $Lu_2Fe_{16}X$ depend on a kind of X-element and generally, they are several times smaller than those in Lu_2Fe_{17}. Magnetocrystalline anisotropy of Lu_2Fe_{17} is strongly reduced by the substitution of Cr for Fe only.

Keywords: Rare-earth intermetallics; Lu_2Fe_{17}; Magnetic properties; High pressure

1. Introduction

The Lu_2Fe_{17} intermetallic compound is an extraordinary member of the family of R_2Fe_{17} intermetallics (R, rare earth). The non-magnetic Lu-element and small volume of crystal cell cause that Lu_2Fe_{17} stays on a verge of stability of ferromagnetism. A presence of both the positive and the negative exchange interactions in Lu_2Fe_{17} intermetallic compound gives rise to complex arrangement of magnetic Fe-moments. The magnetic structure below the Néel temperature, $T_N = 278$ K, was described by helical propagation of ferromagneticaly ordered basal planes along *c*-axis of hexagonal crystal lattice (space group $P6_3/mmc$) [1,2]. The magnetic cell is incommensurate with the crystal lattice. Lu_2Fe_{17} exhibits metamagnetic behaviour below T_N down to a transition temperature, $\Theta_C \sim 200$ K, where spontaneous magnetization occurs. The ferromagnetic arrangement and value of the transition temperature Θ_C are very sensitive to a sample preparation [3]. However, the ferromagnetic ground state of Lu_2Fe_{17} was confirmed by neutron diffraction experiments at ambient pressure [4]. Recently, the ferromagnetic state of Lu_2Fe_{17} was totally suppressed and the antiferromagnetic helical structure was stabilized down to the lowest temperatures under external hydrostatic pressure. The critical pressure P_C is relatively low, $P_C = 0.35$ GPa [4–6]. The metamagnetic transition from the helical back to the ferromagnetic structure can be induced in Lu_2Fe_{17} by an external magnetic field applied in the basal crystal plane. The critical field H_C of this transition increases with increasing pressure. This clearly indicates an increase of the antiferromagnetic interaction between the ferromagneticaly ordered basal planes of Lu_2Fe_{17} under pressure [5]. A presence of the helical and the ferromagnetic structure in Lu_2Fe_{17} was detected by neutron diffraction and magnetization measurements under pressure [4,5]. The observed extreme sensitivity of magnetization, metamagnetic transitions and magnetic arrangement to external conditions drew an attention to the general problems of a volume instability and a non-collinear arrangement of the Fe-moments in all the Fe-rich materials. To bring new information, we infringed a subtle balance of magnetic interactions in Lu_2Fe_{17} by a partial substitution in the Fe sublattice.

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Table 1
Magnetic and magnetoelastic properties of the $Lu_2Fe_{16}X$ intermetallics

	Lu_2Fe_{17}	$Lu_2Fe_{16}Ni$	$Lu_2Fe_{16}Cr$	$Lu_2Fe_{16}Si$
Composition	1.81:17.2	1.64:16.6:0.80	1.61:16.4:0.96	1.69:16.2:1.06
T_C (K)	278[a]	377	377	395
$M_S\|_{5K}$ (μ_B/f.u.)	34.2	33.9	27.7	30.8
$\mu_0 H_A\|_{5K}$ (T)	3.85	3.34	2.7	3.3
$K_1\|_{5K}$ (MJ m^{-3})	−2.48	−2.43	−1.28	−1.96
$K_2\|_{5K}$ (MJ m^{-3})	0.44	0.23	0.11	–
dT_C/dP (K GPa^{-1})	−35[a]	−40	−33	–
$d\ln M_S/dP$ (GPa^{-1})	−0.173[b]	−0.077	−0.048	−0.038
$d\ln K_1/dP$ (GPa^{-1})	−0.54[b]	−0.26	−0.30	–

[a] T_N in Lu_2Fe_{17}.
[b] The values were determined under pressures $P < P_C$ where Lu_2Fe_{17} is ferromagnetic at 5 K.

In this article, pronounced effects of a partial substitution of Ni, Cr and Si for Fe on the magnetic and magnetoelastic properties of the mother Lu_2Fe_{17} intermetallic compound are collected.

2. Experimental

The single crystals of the $Lu_2Fe_{16}X$ (X = Fe, Ni, Cr, Si) intermetallics were prepared by Czochralski method in a tetra-arc furnace under an argon atmosphere. A single crystal state was checked and the crystals were oriented by the X-ray Laue method. The final composition was determined by microprobe analysis on a JEOL electron microscope. Results of the analyses are presented in Table 1. Magnetic properties of Lu_2Fe_{17} at ambient pressure were presented elsewhere [3,7]. The magnetoelastic phenomena were studied in temperature range from 5 to 390 K under hydrostatic pressure up to 1.2 GPa using the miniature high pressure CuBe cell in a SQUID magnetometer. Measured magnetization isotherms were corrected to the internal field $H_i = H_e - NM$, where N is the demagnetizing factor and M is magnetization. Magnetocrystalline anisotropy coefficients K_1, K_2 and anisotropy field H_A of the oriented single crystals were determined by the Sucksmith–Thompson relation [8].

3. Results and discussion

The antiferromagnetic state that was observed in Lu_2Fe_{17} is suppressed and the ferromagnetic arrangement in $Lu_2Fe_{16}X$ is stabilized by the substitution of Ni, Cr and Si for Fe. The Curie temperatures T_C of all the $Lu_2Fe_{16}X$ intermetallics (in Table 1) are about 100 K higher than the Néel temperature of Lu_2Fe_{17}, without a respect to the volume changes induced by substitution (e.g., $\Delta V/V = -0.3\%$ by the Si-substitution [7]). Thus, this significant increase of the ordering temperature cannot be explained by a magnetovolume effect. A substantial decrease of T_C under pressure in $Lu_2Fe_{16}Ni$ and $Lu_2Fe_{16}Cr$ is of the same magnitude as the pressure decrease of the Néel temperature T_N of Lu_2Fe_{17} in pressure range below P_C [6], see Table 1. Moreover, this pressure effect is also well comparable with an effect of hydrogenation. Taking into account a pronounced anisotropy in changes of lattice parameters induced by pressure ($|dc/dP| \gg |da/dP|$) in Lu_2Fe_{17} [6] and by hydrogenation ($da/dx \gg dc/dx$) in $Lu_2Fe_{17}H_x$ [9], we have received the unique value of the Grüneissen parameter $\Gamma = d\ln T_C/d\ln V = 12.5 \pm 0.5$ on the assumption that $d\ln V = 3 \times d\ln a$. The magnetoelastic phenomena in a vicinity of the ordering temperatures T_C and T_N of $Lu_2Fe_{16}X$ intermetallics reflect mainly changes of the exchange interactions in the basal planes that are induced by a variation of the lattice a-parameter. On the other hand, the substituted Ni-, Cr-, Si-atoms exhibit a strong influence on both the intra- and the inter-plane exchange interactions. No indices of a presence of the non-collinear magnetic structures were registered on the magnetization isotherms of the $Lu_2Fe_{16}X$ (X = Ni, Cr, Si) intermetallics that were measured at temperature range from 5 up to 300 K under pressures up to 1 GPa. This can be seen, e.g., in Fig. 1 where the metamagnetic transition is clearly seen on the 5 K isotherm of Lu_2Fe_{17} under pressure only. Almost identical values of Fe-moment, $m_{Fe} = 1.99$; 2.01 and 1.98 μ_B/at.Fe, can be

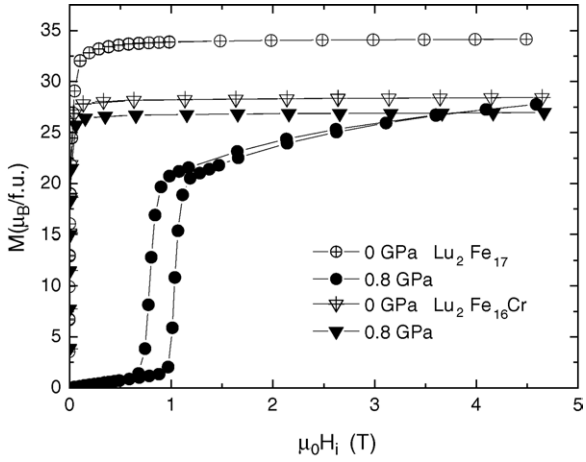

Fig. 1. Magnetization isotherms of Lu_2Fe_{17} (circles) and $Lu_2Fe_{16}Cr$ (triangles) intermetallics at 5 K under ambient and high pressure 0.8 GPa (full symbols).

deduced from the saturated magnetization M_S (5 K, 0) of the Lu_2Fe_{17}; $Lu_2Fe_{16}Ni$ and $Lu_2Fe_{16}Cr$ intermetallics, respectively, on the assumption that $m_{Lu} = 0$, $m_{Ni} = 0.6\ \mu_B$/at.Ni is parallel and $m_{Cr} = 5.0\ \mu_B$/at.Cr is antiparallel with respect to Fe-moments. A smaller Fe-moment, $m_{Fe} = 1.90\ \mu_B$/at.Fe, was observed in the case of the $Lu_2Fe_{16}Si$ intermetallics. The Si-atoms preferentially occupy the 6*g* and 12*k* inequivalent Fe-positions (in the basal planes without Lu atoms) in the substituted compound [7]. This points to a hybridization of Fe- and Si-electron states that can cause the observed decrease of M_S of the $Lu_2Fe_{16}Si$ intermetallics.

The saturated magnetization M_S (5 K, *P*) of Lu_2Fe_{17} has been measured in pressure range below P_C, where the ferromagnetic structure of Lu_2Fe_{17} was verified by neutron diffraction. The observed huge decrease of M_S (see Table 1) can be classified as the Invar anomaly that is regularly accompanied by a large positive spontaneous magnetostriction. The Invar anomalies are ascribed to a subtle balance in occupation of non-bonding and anti-bonding states in vicinity of the Fermi level that results in a volume instability of Fe-moment [10]. Recent calculations of the electron structure of the Y_2Fe_{17} intermetallics revealed that its basic features are identical with ones of the electron structure of the fcc-FeNi Invar alloys [11]. So, we are competent to characterize the observed great values of $d\ln M_S/dP$ in the case of the Lu_2Fe_{17} intermetallics as the Invar anomaly using this model. From this point of view, the substitution of Ni, Cr and Si for Fe in Lu_2Fe_{17} can be considered as a breaking of the subtle balance mentioned above that leads to a more stable Fe-moment, to the increase of T_C and to a weakening of the magnetoelastic phenomena illustrated by reduced values of $d\ln M_S/dP$ of the $Lu_2Fe_{16}X$ intermetallics. A comparison of the pressure dependence of M_S (5 K) of Lu_2Fe_{17} and $Lu_2Fe_{16}Cr$ is presented in Fig. 1. Temperature dependence of magnetization, M_S (*T*, *P*), is identical (in relative values) in both the Lu_2Fe_{17} and the $Lu_2Fe_{16}Ni$ intermetallics, see Fig. 2. Fig. 2 illustrates simultaneously the difference in the pressure decrease of M_S in the mentioned intermetallics. Values of $d\ln M_S/dP$ for all the $Lu_2Fe_{16}X$ (X = Ni, Cr, Si) intermetallics are collected in Table 1.

The magnetocrystalline anisotropy of Lu_2Fe_{17} is only slightly affected by the substitution that does not change the crystal symmetry and the crystal electric field (CEF) interactions. The anisotropy field H_A is almost identical in the Lu_2Fe_{17}, $Lu_2Fe_{16}Ni$ and $Lu_2Fe_{16}Si$ intermetallics ($\mu_0 H_A = 3.6 \pm 0.25$ T). A negligible effect of hydrogenation on magnetocrystalline anisotropy of the $Lu_2Fe_{17}H_x$ hydrides was presented in Ref. [9]. A significantly lower value of H_A has been observed in the case of the $Lu_2Fe_{16}Cr$ intermetallics only. However, the lower value of H_A of this intermetallic compound remains proportional to the value of its saturated magnetization M_S as it can be seen in Table 1 for all the studied intermetallics. A temperature dependence of H_A of $Lu_2Fe_{16}Cr$ exhibits a rather different character in comparison with one of $Lu_2Fe_{16}Ni$, see Fig. 3. On the other hand, H_A decreases substantially with pressure in all the studied intermetallics. The very high values of $d\ln K_1/dP|_{5K}$ (in Table 1) reflect a dependence of the magnetocrystalline anisotropy on both the CEF interactions and the magnetization. The lattice *c*-parameter decreases with pressure more significantly than the *a*, *b*-parameters in basal plane. A decrease of the *c*/*a* parameter under pressure induces a variation of the CEF interactions that cause together with the decrease of magnetization under pressure a high sensitivity of the magnetocrystalline anisotropy of all the $Lu_2Fe_{16}X$ (X = Ni, Cr, Si) intermetallics to the external pressure.

No antiferromagnetic arrangement of Fe-moments has been indicated in the $Lu_2Fe_{16}X$ (X = Ni, Cr, Si) intermetallics. The ferromagnetic state is stabilized and the huge magnetoelastic phenomena observed in the Lu_2Fe_{17} intermetallics are reduced in the $Lu_2Fe_{16}X$ intermetallics by the substitution of Ni, Cr and Si for Fe. The study of the substi-

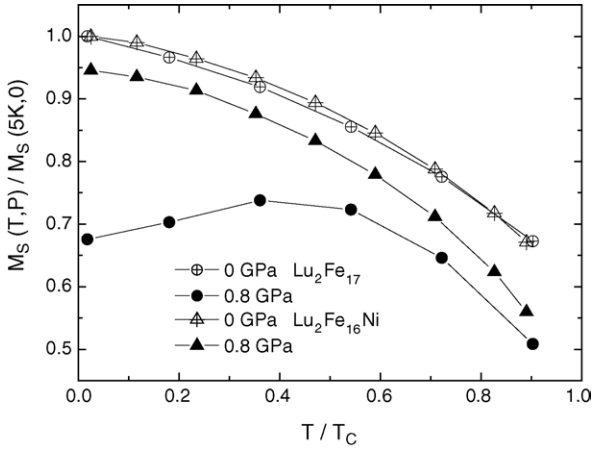

Fig. 2. Temperature dependencies of saturated magnetization of Lu_2Fe_{17} (circles) and $Lu_2Fe_{16}Ni$ (triangles) intermetallics under ambient and high pressure 0.8 GPa (full symbols).

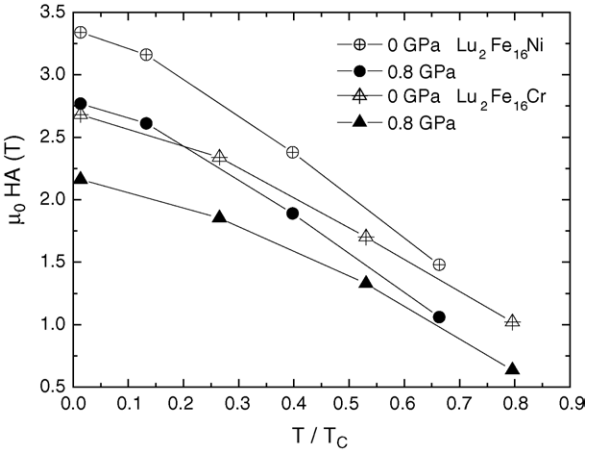

Fig. 3. Temperature dependencies of anisotropy field of $Lu_2Fe_{16}Ni$ (circles) and $Lu_2Fe_{16}Cr$ (triangles) intermetallics under ambient and high pressure 0.8 GPa (full symbols).

tuted intermetallics has pointed to the important role of the anisotropy of magnetoelastic effects in the magnetic behavior of the R–Fe intermetallics.

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