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Spontaneous Magnetostriction of $Lu_2Fe_{17-x}Si_x$

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Thermal expansion of $Lu_2Fe_{17-x}Si_x$ solid solutions was measured by X-ray powder diffraction. The magnetic ordering in all compounds within the homogeneity range ($x \leq 3.4$) is accompanied by a large spontaneous volume magnetostriction distributed anisotropically over the principal axes of the hexagonal crystal structure. The volume effect ω_s in the ground state reaches 14.7×10^{-3} in Lu_2Fe_{17} and decreases monotonously to 8.9×10^{-3} at $x = 3.4$ following the reduction of magnetic moment. Despite still large ω_s, the Invar behavior observed in Lu_2Fe_{17} changes to a positive thermal expansion for $x > 1$ due to increasing Curie temperature.

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1. Introduction

R_2Fe_{17} intermetallic compounds are known to undergo large spontaneous magnetostrictive deformations originated mainly from the Fe sublattice [1–3]. This leads, in combination with relatively low (for so high Fe content) Curie temperatures T_C, to Invar behavior in a wide temperature range. Many investigations have shown that T_C of R_2Fe_{17} can be increased considerably by substitution of a variety of other elements on the Fe sites [4]. The substitution (or interstitial introduction by light elements) modifies also the magnetoelastic properties and their influence on the thermal expansion [5].

In the present work, we studied the thermal expansion anomalies accompanying the magnetic ordering and determined the linear and volume spontaneous magnetostrictions in $Lu_2Fe_{17-x}Si_x$. In these solid solutions on the base of Lu_2Fe_{17} with non-magnetic Lu, the Si solubility extends up to $x = 3.4$ [6]. The antiferromagnetism observed in Lu_2Fe_{17} at elevated temperatures (in the ground state Lu_2Fe_{17} is ferromagnetic) is suppressed, all the solid solutions are ferromagnetic and T_C increases up to 485 K at the solubility limit. The Fe magnetic moment and the anisotropy energy decrease with Si substitution.

2. Experimental

The powder samples of compounds with $x = 0$, 1, 2, and 3.4 were prepared from parts of single crystals studied in Ref. [6]. The measurements were carried out on a Siemens D-500 diffractometer equipped with a helium-flow cryostat (Oxford Instruments CF1108T) in the Bragg–Brentano geometry using filtered cobalt radiation (Co K_α) in the temperature range 5–500 K. Extension of the measurements to 600 K was performed in a home-made high-temperature chamber. The data reduction was performed using the Rietveld analysis [7] (the computer program Fullprof [8] was employed for the refinement).

3. Results and discussion

Temperature dependences of the lattice parameters a, c, and the unit-cell volume V are presented in Figs. 1 and 2, respectively, whereas Fig. 3 shows the Si-concentration dependence of the magnetic and magnetoelastic properties. All the compounds crystallize in the hexagonal crystal structure of the Th_2Ni_{17} type. The Si atoms substitute the Fe ones on the $6g$ and $12k$ positions, the other Fe sites are Si-free. The lattice parameters and the unit-cell volume decrease with increasing Si content (Fig. 3).

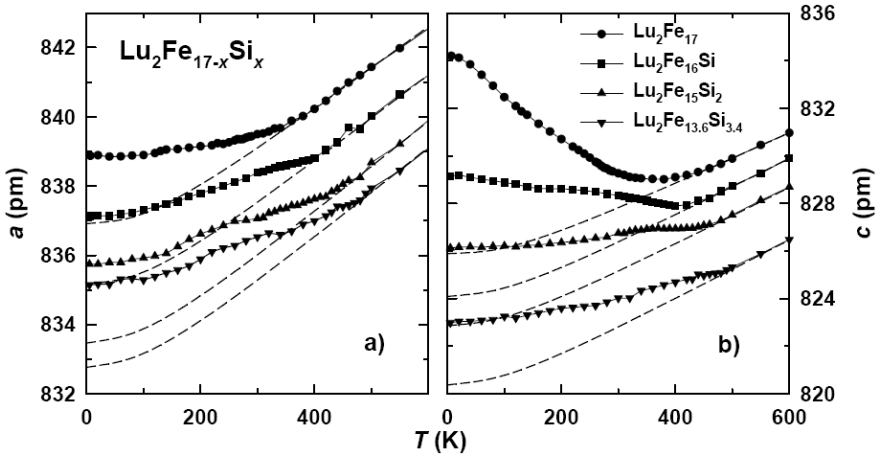

Fig. 1. (a) Temperature dependence of the lattice parameters a for $Lu_2Fe_{17-x}Si_x$. The dashed lines represent the extrapolation of the $a(T)$ dependence from the paramagnetic to the ferromagnetic range using a Debye temperature Θ_D of 450 K. (b) The same for parameter c.

The experimental temperature dependences of the lattice parameters in the paramagnetic range were extrapolated to the magnetically-ordered range using Debye temperature value 450 K determined from acoustic measurements of R_2Fe_{17} [3]. (For details of the extrapolation procedure, see Ref. [3].)

The relative difference between the experimental and the extrapolated values of the lattice parameters are the spontaneous linear magnetostrictive strains λ_a and λ_c. The temperature dependences of the volume spontaneous magnetostriction $\omega_s = 2\lambda_a + \lambda_c$ are shown in Fig. 2b.

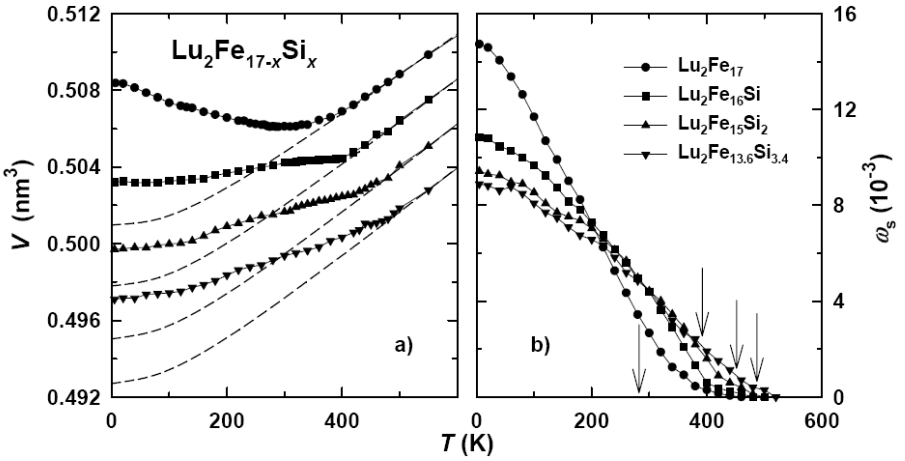

Fig. 2. Temperature dependence of the unit-cell volume V (a) and volume spontaneous magnetostiction ω_s (b). The arrows indicate the ordering temperature values.

The easy-plane linear spontaneous magnetostriction λ_a depends on the Si content only slightly and in all compounds studied is not high enough to overcome the phonon thermal-expansion contribution and provide the Invar-like behavior (Fig. 1a). λ_c along the c axis is considerably larger and the negative thermal expansion is observed for $x = 0$ and 1 (Fig. 1b). As regards the unit-cell volume, only Lu_2Fe_{17} exhibits the Invar properties due to huge $\omega_s = 1.5\%$ and relatively low ordering temperature. Let us note that this compound orders antiferromagnetically at $T_N = 278$ K and undergoes a transition to the ferromagnetic state at T_C about 140 K (all other samples are ferromagnets in the whole range of the magnetic ordering). The absence of a noticeable anomaly at T_C in Lu_2Fe_{17} indicates that magnetoelastic effects in this compound originate from the ferromagnetic Fe–Fe exchange interactions within the basal plane, the inter-plane antiferromagnetism does not influence the magnetoelasticity.

All the compounds have the easy-plane magnetic anisotropy and therefore should have an orthorhombic magnetostrictive distortion. However, no distortion was observed which means that the anisotropic magnetostriction responsible for the distortion is below the experimental sensitivity 2×10^{-4}, e.g. at least by 1 order lower than the exchange magnetostriction seen in the volume effect 0.9–1.5%.

One can see in Fig. 2b that ω_s in Lu_2Fe_{17} does not vanish at T_N but, as in other R_2Fe_{17} [3], persists at considerably higher temperatures indicating a wide region of short range magnetic order. Stabilization of the ferromagnetism upon Si

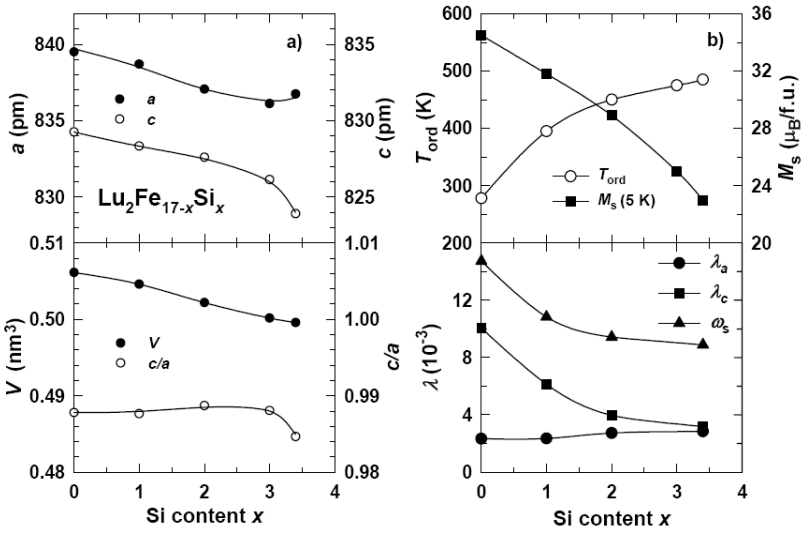

Fig. 3. Concentration dependence of the lattice parameters a and c, ratio c/a, and unit-cell volume V (panel a) and of the ordering temperature T_C (T_N at $x = 0$), spontaneous magnetic moment M_s, linear spontaneous magnetostrictive strains λ_a, λ_c, and volume spontaneous magnetostriction ω_s (panel b) in $Lu_2Fe_{17-x}Si_x$ solid solutions.

substitution reduces this range and the temperature, where ω_s disappears, corresponds well to T_C.

Acknowledgments

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