$(Lu_{0.8}Ce_{0.2})_2Fe_{17}$のメタ磁性転移における強磁場磁気歪みの観測
Magnetostriction at the metamagnetic phase transition in $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$

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Abstract

The antiferromagnetic state, observed in Lu_2Fe_{17} at elevated temperatures, is stabilized in $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ over whole magnetically ordered range below T_N = 247 K. The compound exhibits easy-plane magnetic anisotropy and a metamagnetic transition at fields below 1 T. The magnetostriction at the metamagnetic transition was studied by both bulk (the capacitor method on a single crystal) and microscopic (X-ray powder diffraction) methods. The results are complementary and show a lattice expansion both in the basal plane and along the *c*-axis with a very large volume effect up to 0.6%. Negative thermal expansion along the *c* axis is observed up to room temperature, far above T_N. This points to an extended temperature interval of short-range order.

1. Introduction

The very short Fe-Fe distances d_{FeFe} between Fe atoms at some positions are special features of the rare-earth intermetallic compounds R_2Fe_{17}. A negative exchange interaction between Fe atoms coexists with positive interactions in the case of larger d_{FeFe} distances between Fe atoms at other positions. The total exchange interaction is positive, however, existence of these competitive interactions leads to unexpectedly low Curie temperatures (T_C) in compounds with such a high content of Fe. The compound $Lu_2Fe_{17}$with the smallest non-magnetic rare-earth element and an hexagonal crystal structure of the Th_2Ni_{17} type exhibits an incommensurate helimagnetic structure below the Néel temperature T_N = 274 K [1,2]. The length of the magnetic helix increases continuously with decreasing temperature and a transition to a ferromagnetic phase occurs below 130 K. The delicate balance of ferro- (F) and antiferro- (AF) magnetic interactions is very sensitive to changes of the external conditions. Therefore, Lu_2Fe_{17} is a very apt compound to study the interplay of magnetic field, temperature, external pressure and substitution effects on the magnetism of high-Fe-content intermetallics. In $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$, the AF arrangement of the magnetic moments is stabilized down to the ground state [3]. The compound has easy-plane magnetic anisotropy. Figure 1 shows that magnetization isotherms along the *a*-axis of $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ exhibit the metamagnetic transition. It can be concluded that the transition is of the first order due to a wide field hysteresis.

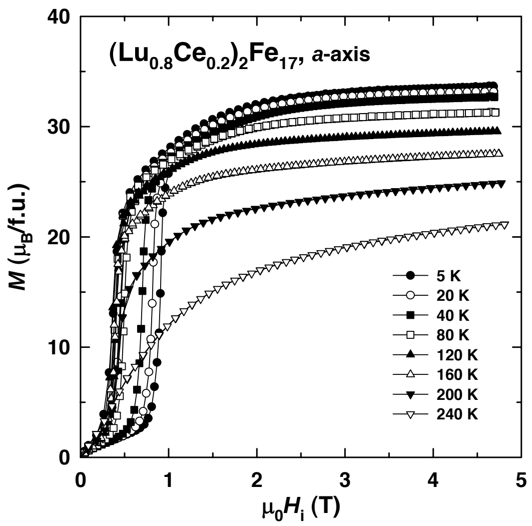

Fig. 1. Magnetization curves of $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ along the *a* axis at different temperatures.

The magnetization does not saturate immediately after the transition but reaches its saturation value of 35 μ_B at about 1 T above the transition field. We suppose that a first-order transition from the AF to a non-collinear F state occurs, followed by gradual alignment of the magnetic moments to the collinear F state. Both stages of the magnetization process are expected to exhibit a pronounced magnetostriction.

In the present study, we report on the results of magnetostriction measurements performed by bulk and microscopic (X-ray powder diffraction) methods.

2. Experimental

The single crystal of $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ was prepared by the Czochralski method in a tetra-arc furnace. The magnetic isotherms were measured in Prague in a PPMS-9 magnetometer (Quantum Design) along the principal axes of the single crystal and on an isotropic powder sample in fields of up to 9 T in the 5-300 K temperature range and in Sendai in a superconducting quantum interference device (SQUID) magnetometer on a powder sample in the 5-300 K temperature interval in fields of up to 5 T.

The longitudinal and transverse magnetostriction were measured on the $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ single crystal in the PPMS-9 in Prague by the capacitor method along the *a* and *c* axes in fields applied along the *a* axis. High-field powder X-ray diffraction experiments were carried out in Sendai with CuKα radiation at 10-300 K using a Gifford-McMahon (GM) type cryocooler (helium gas closed-cycle refrigerator) and for $B \leq 5$ T using a cryocooled split-pair superconducting magnet [4]. A powder sample was fixed by means of Apiezon grease on a copper-boat holder, which was attached to the second stage of the GM cryocooler in the cryostat. We checked that the powder sample was not removed from the sample holder by the magnetic force. The diffraction data were obtained for $20° \leq 2\theta \leq 100°$ with step of 0.01°.

3. Results and discussion

We measured the magnetostriction by two independent methods because we wanted to combine the high sensitivity and relative accuracy of the capacitor method with X-ray dilatometry, which has a much lower sensitivity and accuracy (~10^{-4}) but is a direct method of determination of the interatomic distance changes. Since we expected the linear magnetostriction strains at the metamagnetic transition to exceed 10^{-3}, the X-ray dilatomery was considered to indicate and measure such effects rather well. In the case of other types of dilatometry, an ambiguous point is the calculation of the volume effect from the linear strains, because zero-field state should be exactly the same for measurement of each linear effect, which is difficult to be arranged and controlled. Furthermore, possible field-induced structure changes can be observed by means of X-ray diffraction.

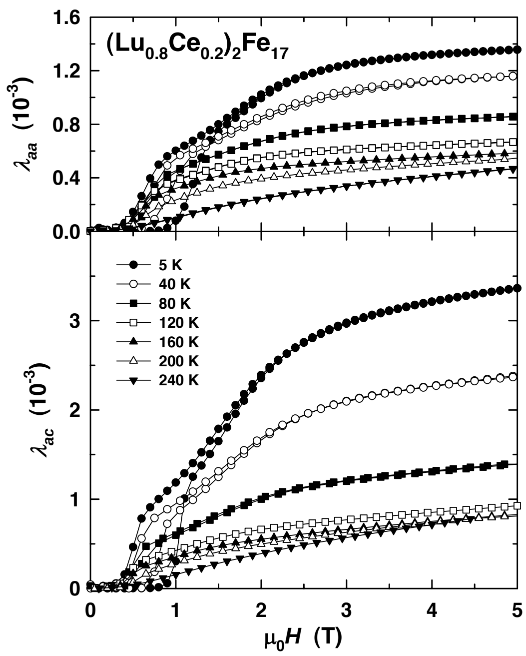

Fig. 2. Dependence of the longitudinal (top) and transverse (bottom) magnetostriction of $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ on a magnetic field applied along the *a*-axis at different temperatures.

Figure 2 presents the results of the bulk measurements. One can see that the metamagnetic transition is accompanied by a large lattice expansion both along the *c*-axis and in the basal plane. Comparison with the magnetization curves (Fig. 1) shows that the second stage of the transition from the AF to F state, i.e., after the magnetization jump up to the saturation, influences the interatomic distances even stronger than the first stage.

The field evolution of the (600) and (306) reflections at 10 K are shown in Fig. 3. With increasing field, both lines shift towards smaller 2Θ, indicating expansion of the lattice. The α_1-α_2 doublet is well-resolved at low and high fields whereas, between 0.5 and 3.5 T, the line profiles are characteristic for the coexistence of two phases and agree well with the observed hysteresis in the magnetization. The second-order magnetization process without hysteresis at 200 K does not change the line profiles, as seen in Fig. 4.

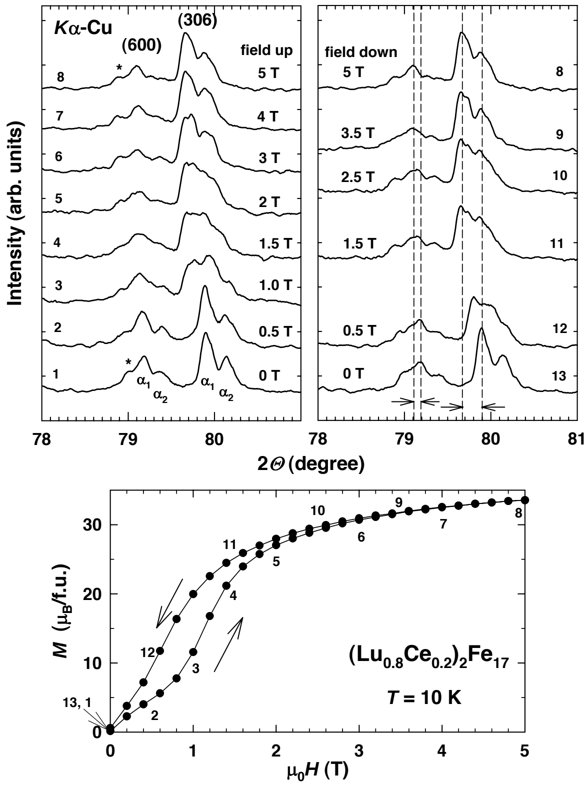

Fig. 3. Magnetization curve and hysteresis loop of a fixed random powder sample of $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ and corresponding profiles of the (600) and (306) reflections in Cu$K\alpha$ radiation at 10 K. The numbers of the profiles correspond to the ones on the magnetization curve. The dashed lines indicate the position of the α_1-(600) and α_1-(306) reflections at 0 and 5 T.

* indicates a reflection of unknown origin.

In Fig. 5 we compare the results obtained by the two methods. Since the samples were different (single- and polycrystals), and the magnetic anisotropy is strong at low temperatures, we can compare only the results of linear magnetostriction along the *a* (λ_a) and *c*-axis (λ_a) in high fields because here both samples are magnetically saturated. At 200 K, where the anisotropy is weak, we can also compare the results at lower fields. Taking into account the large error of X-ray dilatometry (10^{-4}), we may conclude that the agreement is rather good.

Since $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ has easy-plane type of magnetic anisotropy, it should undergo an orthorhombic magnetostrictive distortion at magnetic ordering, especially at low temperatures. If the distortion would be large, we need to measure the magnetostriction in one more geometry in addition to the one presented in Fig. 2 - field along *a*, strain along [120] (*b*-axis in the orthorhombic notation) - in order to determine the volume effect. The X-ray study revealed no distortion which means that the anisotropic magnetostriction, responsible for the distortion, is below the experimental sensitivity $1*10^{-4}$, i.e. much smaller than the observed values in Fig. 2. Therefore, we can neglect such distortion and determine the volume effect as $\omega = 2\lambda_a + \lambda_c$. The results of $\omega(H)$ are presented in Fig. 5 as well.

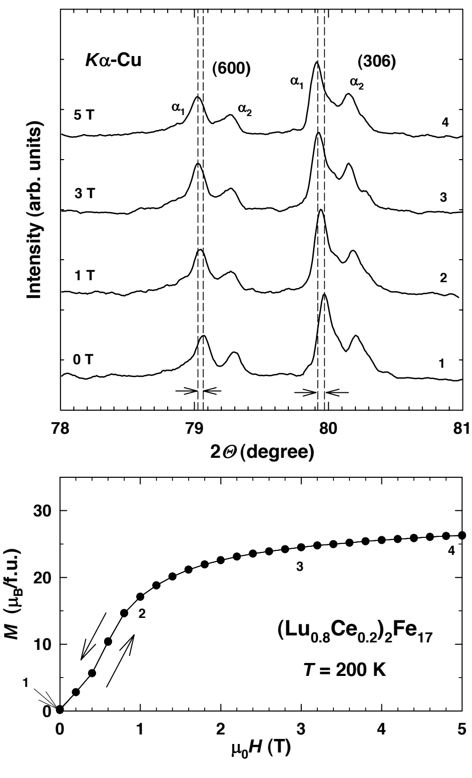

Fig. 4. The same as Fig. 3, but at T = 200 K. No hysteresis and no two-phase state are observed.

At 5 T, the volume effect reaches a very large value of 0.6%. This is comparable with the volume spontaneous magnetostriction ω_s = 1.5% in Lu_2Fe_{17} [5] and 1% in Ce_2Fe_{17} [6]. However, the latter is the volume difference between the paramagnetic and the ordered state whereas the present results correspond to an order-order transition which may be expected to have a smaller volume effect.

Usually, the magnetostriction is proportional to the square of the magnetization. However, in $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$, $\omega(T)$ decreases much faster with increasing temperature than M^2 as seen in Fig. 6 where the dashed line represents the fit $\omega(T) = \omega(0) [M(T)/M(0)]^2$ in which for $\omega(0)$ the experimental low-temperature value has been taken. The slope of $\omega(T)$ dependence changes in the vicinity of 150 K. In the same temperature range 140-160 K, the magnetization hysteresis disappears and the first-order metamagnetic transition is replaced by a S-shape magnetization curve. The temperature dependence of the transition field H_c (determined from the inflection point of the $M(H)$ curves) also changes its character at the same temperatures (see the inset in Fig. 6).

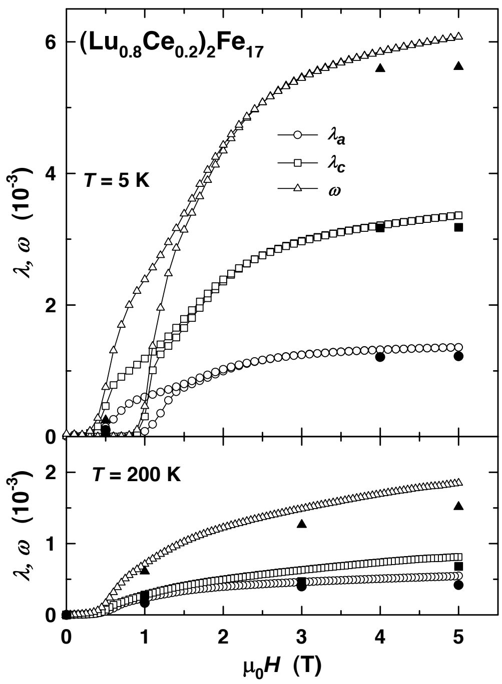

Fig. 5. Comparison of the magnetostriction curves measured along the a and c axes of a $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ single crystal in a field applied along the a axis at 5 K and 200 K (open symbols) with results obtained by x-ray powder diffraction on fixed random powder at 10 K and 200 K, (filled symbols). $\omega = 2\lambda_a + \lambda_c$ represents the volume magnetostriction

All these features point to a possible phase transition similar to the one observed in Ce_2Fe_{17} at 124 K [7] between two antiferromagnetic phases. This transition is accompanied by the minor structure changes and a noticeable effect in the temperature dependence of specific heat $C_p(T)$. In the case of $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$, neither structural changes nor peak in $C_p(T)$ are observed, which might be due to atomic disorder, an intrinsic feature of quasibinary compounds. Due to inhomogeneities on an atomic scale, the transition in $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ is spread over a wide temperature interval whereas in the binary Ce_2Fe_{17}, it is sharp.

The lattice parameter c exhibits a negative thermal expansion not only within the magnetically ordered state below T_N = 247 K but also up to room temperature indicating a wide temperature interval of short-range magnetic order. Such behavior is typical for R_2Fe_{17} compounds, e.g., in Lu_2Fe_{17} with T_N = 274 K, a deviation from the paramagnetic $c(T)$ extends up to 400 K [5]. The presence of such a wide temperature range of short-range order did not allow us to determine the spontaneous magnetostriction in $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$. Extension of the X-ray diffraction measurements up to at least 500 K is in progress.

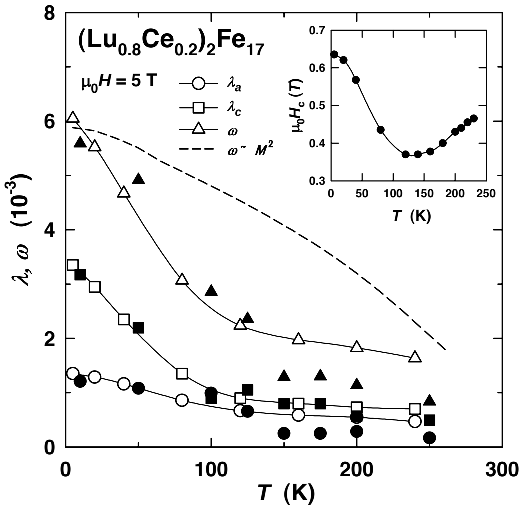

Fig. 6. Temperature dependence of the linear and volume magnetostriction of $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ in 5 T, measured by the capacitor method on single crystal (field along the a-axis) (open symbols) and by X-ray diffraction on a fixed random powder sample (filled symbols). The dashed line is the $\omega(T) \sim M^2(T)$ fit (see text). The inset shows the temperature dependence of the critical field of the metamagnetic transition.

4. Conclusions

A magnetostriction study of $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ has been performed by two methods - bulk measurements (the capacitor method on a single crystal) and X-ray powder diffraction. $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ is an antiferromagnet below T_N = 247 K. It exhibits easy-plane type of magnetic anisotropy and a metamagnetic transition at fields below 1 T, followed by alignment of the non-collinear ferromagnetic structure into a field-induced collinear ferromagnet.

The field-induced transitions are accompanied by lattice expansion both in the basal plane and along the *c*-axis. At low temperatures, the linear expansion along the *a*- and *c*-axis and the volume effect reach $\lambda_a = 1*10^{-3}$, $\lambda_c = 3.8*10^{-3}$ and $\omega = 5.8*10^{-3}$, respectively.

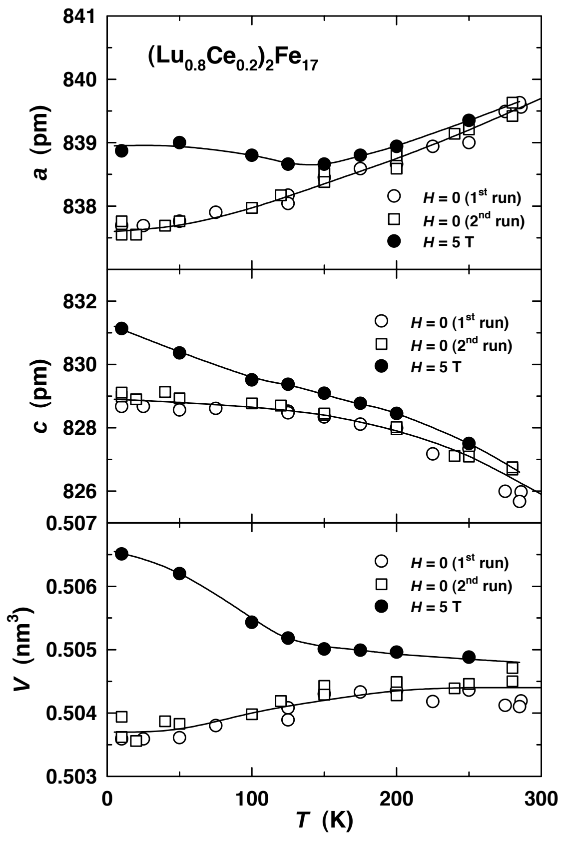

Fig. 7. Temperature dependence of the lattice parameters *a* and *c* and the unit-cell volume *V* of $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$ in 0 and 5 T.

With increasing temperature, the magnetostriction rapidly decreases. Its temperature dependence becomes smooth above about 150 K. Along with the other observations, this may indicate the presence of an order-order transition between two antiferromagnetic phases which is broadened due to inhomogeneities on an atomic scale.

A negative thermal expansion along the *c* axis is observed far above T_N which indicates an extended temperature interval of the short-range order. This has prevented us to derive the spontaneous magnetostriction from the temperature dependencies of the lattice parameters. Extension of the X-ray diffraction measurements up to 500 K is currently in progress in order to complete the magnetostriction study of $(Lu_{0.8}Ce_{0.2})_2Fe_{17}$.

Acknowledgements

The work is a part of the research project AVOZ10100520 and has been supported by grants GACR 202/06/0185 and GAUK 109-10/257015. The high-field X-ray diffraction measurements were carried out at the High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University. The magnetization measurements were performed in the SQUID magnetometer at the Center for Low Temperature Science, Tohoku University.

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