$Y_2Fe_{17-x}Ru_x$の磁気状態と磁歪
Magnetic states and magnetostriction in $Y_2Fe_{17-x}Ru_x$ system

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1. Introduction

Among various Fe-containing compounds exhibiting the Invar effect, alloys with a high Fe content R_2Fe_{17} (R is the rare earth metal or Y) deserve attention for the peculiar magnetism and pronounced magnetoelastic properties [1]. The distinctive feature of R_2Fe_{17}, having the anomalously low ordering temperatures across the series, is a coexistence of positive and negative exchange interactions between Fe atoms at different crystallographic sites of the rhombohedral Th_2Zn_{17} (for the light R) and hexagonal Th_2Ni_{17} (for the heavy R) crystal structures (the compounds with R = Y can be formed in both structural modifications) [2]. The main contribution into the negative exchange comes from the electron hopping at the very short dumbbell-like Fe pairs (6*c*/4*f* Wyckoff positions in the rhombohedral and hexagonal structures, respectively). The magnetic structures of R_2Fe_{17} are ferro- or ferrimagnetic depending on the type of the rare earth except for the compounds with R = Ce and Lu, which exhibit the non-collinear magnetism within the certain temperature ranges.

The magnetic properties of R_2Fe_{17} can be greatly and rather easily influenced by substitutions, applied pressure or by introduction of the interstitial atoms [3-5]. Despite the fact that Y_2Fe_{17} is a collinear ferromagnet under normal conditions, destabilization of the collinearity and existence of a noncollinear magnetic structure in Y_2Fe_{17} was found at sufficiently large pressures [4]. In the present work, the investigation of Ru substitution on the ferromagnetic state stability in Y_2Fe_{17} was conducted together with the magnetostriction study on $Y_2Fe_{17-x}Ru_x$ (x = 0, 0.25, 0.5, 0.75, 1) system.

2. Experimental

The single crystals of $Y_2Fe_{17-x}Ru_x$ (x = 0, 0.25, 0.5, 0.75, 1) were prepared by the Czochralski method in a tri-arc furnace under similar conditions previously reported in Ref. [3]. The back-scattering Laue patterns were used to check the monocrystalline state and to orient the crystals for cutting the samples. The magnetic isotherms were measured along the principal axes of the single crystals in fields up to 5 T in the 5-300 K temperature range on a SQUID magnetometer (Quantum Design).

The powder X-ray diffraction (XRD) experiments were carried out in the High Field Laboratory for Superconducting Materials of Tohoku University 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. The diffraction data were obtained for $10° \leq 2\theta \leq 100°$ with the step of 0.01°. The positions of diffraction peaks and the lattice parameters were determined with the use of FullProf refinement package. The samples for XRD were prepared from the single crystals by crushing them into fine powder. For the measurement, the samples were fixed on a copper boat by the Apiezon N grease.

3. Results and discussion

a) Crystal structure of $Y_2Fe_{17-x}Ru_x$

The hexagonal crystal structure of the Th_2Ni_{17} type was approved for all of the $Y_2Fe_{17-x}Ru_x$ (x = 0, 0.25, 0.5, 0.75, 1) compounds with the lattice parameters depicted in Fig. 1. At the lowest Ru concentration, the lattice parameters of $Y_2Fe_{16.75}Ru_{0.25}$ do not differ much from those of the parent Y_2Fe_{17} (nevertheless, drastic difference in magnetic properties of the compounds is shown in the next section). As the Ru content increases, the substitution of Ru for Fe i n $Y_2Fe_{17-x}Ru_x$ results in the unit cell volume increase, however, the c/a ratio remains unchanged upon the Ru concentration rise.

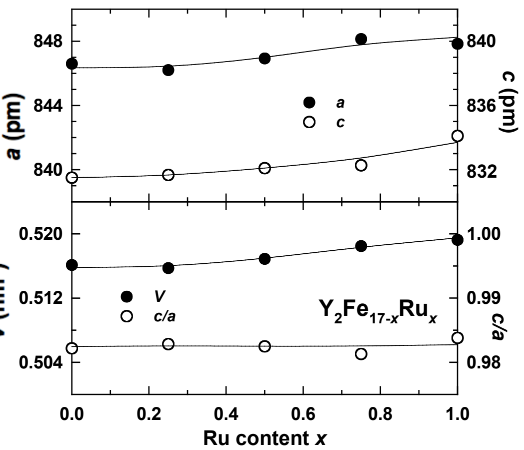

Fig. 1. Ru-concentration dependence of the lattice parameters a, c, the unit-cell volume V and the ratio c/a in $Y_2Fe_{17-x}Ru_x$ system.

In the present work, we did not intend to determine the crystallographic positions occupied by Ru atoms within the Th_2Ni_{17} structure. However, one may assume similar filling scheme for Ru atoms as in the isostructural compound $Lu_2Fe_{16.5}Ru_{0.5}$, where the occupation of the 12k lattice sites by Ru atoms was justified by powder neutron diffraction [6].

b) Magnetization study

Magnetization curves measured along the a-axis for the $Y_2Fe_{17-x}Ru_x$ single crystals at 5 K are presented in Fig. 2. All the compounds have the easy-plane type of magnetic anisotropy with the hard c-axis magnetization direction (the c-axis curves are not displayed here). The alloy of Y_2Fe_{17} shows ferromagnetic behavior with the spontaneous magnetic moment of 35 μ_B/f.u.. Substitution of as much as 0.25 Ru at./f.u. for Fe, is found to induce non-collinearity of magnetic moments in $Y_2Fe_{16.75}Ru_{0.25}$ revealed both in the decrease of spontaneous magnetic moment and in the appearance of small hysteresis in low fields. Most probably, the low-temperature magnetism of $Y_2Fe_{16.75}Ru_{0.25}$ can be described with a fan ferromagnetic structure, which transforms into an antiferromagnetic one at higher temperatures. The temperature dependence of magnetization of $Y_2Fe_{16.75}Ru_{0.25}$ shows a typical peak of the ant iferromagnetic-to-paramagnetic transition at the

The magnetization curves of the $Y_2Fe_{17-x}Ru_x$ Néel point of 263 K.compounds with the Ru content x > 0.25, on

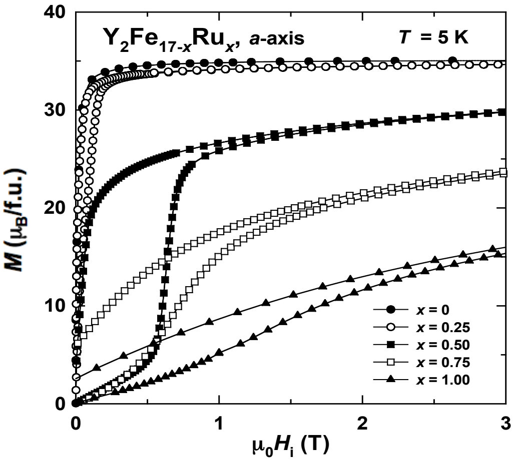

Fig. 2. Magnetization curves along the a-axis of $Y_2Fe_{17-x}Ru_x$ single crystals at 5 K.

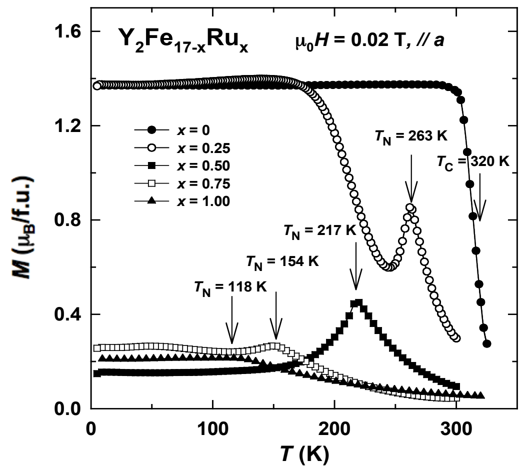

Fig. 3. Temperature dependence of magnetization measured along the a-axis of $Y_2Fe_{17-x}Ru_x$ single crystals in 0.02 T field.

the contrary, do not possess any spontaneous component of magnetization and reveal a very low initial susceptibility with the characteristic hysteretic field-induced transitions above certain critical fields.

Similar metamagnetic behavior was observed previously for the isostructural compounds $Lu_2Fe_{17-x}Ru_x$ with x = 0.5 and 1 [3] (for $Lu_2Fe_{16.5}Ru_{0.5}$, presence of a helimagnetic AF structure was justified by powder neutron diffraction [6]). The magnetic behavior of $Y_2Fe_{17-x}Ru_x$ system with x = 0.5 and 1 reproduces that of

$Lu_2Fe_{17-x}Ru_x$ and, therefore, one may suggest formation of identical, antiferromagnetic structures in both systems $R_2Fe_{17-x}Ru_x$ with Y and Lu. The magnetization process of $Y_2Fe_{16.5}Ru_{0.5}$ consists of two stages: the metamagnetic transition from the non-collinear AF into the non-collinear F state in magnetic fields of 0.7 with a subsequent turn of magnetic moments at higher fields. The magnetization curves for $Y_2Fe_{17-x}Ru_x$ with a highest Ru content do not tend to saturate in the highest applied field, and the metamagnetic transitions along the easy axis occur within a wider field region.

The 4*d*-electrons of Ru (shielded more by the inner core electrons than the Fe 3*d*-electrons) most likely change the occupation of anti- and non-bonding 3*d*-states lying close to the Fermi level that reduces the total magnetic moment in $Y_2Fe_{17-x}Ru_x$ (Fig. 2) with the gradual increase of Ru content. Due to the change of the density of states profile at the Fermi level, altering the competitive local exchange interactions (with strengthening of the negative ones) leads to suppression of the ferromagnetic states and results in the stabilization of the antiferromagnetic ones in the whole range of magnetically ordered state. The ordering temperatures, corresponding to the peaks on $M(T)$ curves shift towards the lower temperatures as the Ru concentration increases (Fig. 3).

c) Magnetostriction in $Y_2Fe_{17-x}Ru_x$ (x = 0.25, 0.5, 0.75, 1)

One can expect to observe pronounced magnetostrictive strains down to the lowest temperatures in $Y_2Fe_{17-x}Ru_x$ system as in the case of the recently studied $Lu_2Fe_{16.5}Ru_{0.5}$ compound, where the field-induced metamagnetic AF-F transitions were manifested in the large lattice expansion along the *c*-axis (λ_c) and in the basal plane (λ_a). The X-ray diffraction patterns were measured in high magnetic field for all the compounds under study. Figure 4 shows the field/temperature evolution of the characteristic (600) and (306) reflections for $Y_2Fe_{16.75}Ru_{0.25}$ and $Y_2Fe_{16.5}Ru_{0.5}$ compounds at 10 K (top) and 120 K (bottom). The

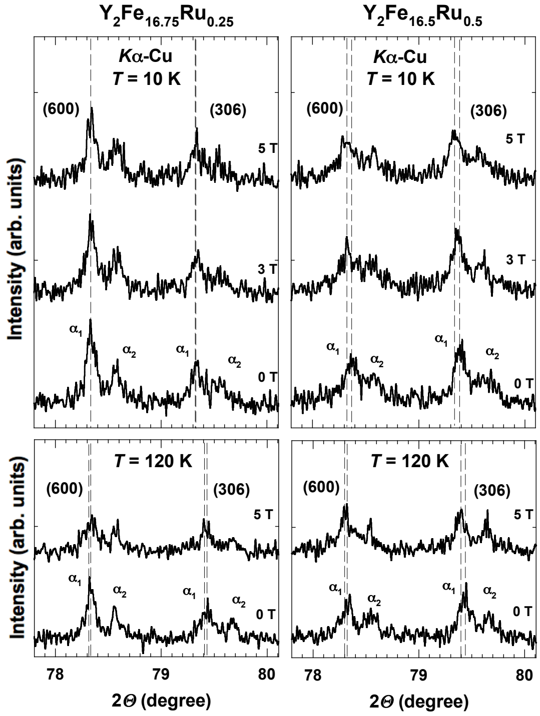

Fig. 4. The (600) and (306) reflection profiles of $Y_2Fe_{16.75}Ru_{0.25}$ (left) and $Y_2Fe_{16.5}Ru_{0.5}$ (right) powder samples measured in Cu*K*α radiation at 10 K (top) and 120 K (bottom) in various fields. The dashed lines indicate the α_1-(600) and α_1-(306) reflections positions at 0 and 5 T.

application of magnetic field at 10 K does not induce the magnetostrictive strains in the $Y_2Fe_{16.75}Ru_{0.25}$ compound, which has a ferromagnetic-fan ground state. However already at 120 K (i.e. still below the antiferromagnetic state), the (600) and (306) peak lines shift towards the smaller angles 2Θ, indicating the expansion of the lattice as the result of expected increase of the angle of a fan structure, when approaching the antiferromagnetic state at higher temperatures. For $Y_2Fe_{16.5}Ru_{0.5}$, pronounced line shift towards the smaller angles 2Θ is observed at 10 K in 5 T field. The α_1-α_2 doublet shape also slightly evolves as the magnetic field increases from 3 to 5 T, i.e. at the field range corresponding to the second stage of a magnetization process (gradual increase of magnetization) after the field-induced metamagnetic transition (see Fig. 2).

Despite the fact that $Y_2Fe_{17-x}Ru_x$ have the easy-plane type of magnetic anisotropy, the volume effect can be determined as $\omega = 2\lambda_a + \lambda_c$ [1] since no orthorhombic magnetostrictive distortion in the basal (no splitting or noticeable broadening of the (600) line between 10 and 120 K in Fig. 4) was observed in the course of the XRD study. The magnetostriction data for the compounds studied is presented in Table 1.

Table 1. Magnetostriction values along the *c*-axis (λ_c) and in the basal plane (λ_a) and the volume effect ω of $Y_2Fe_{17-x}Ru_x$ (x = 0.25, 0.5, 0.75, 1) at 10 K and in 5 T field.

Sample	$\lambda_{a\|5T}$ (10^{-3})	$\lambda_{c\|5T}$ (10^{-3})	$\omega_{\|5T}$ (10^{-3})
$Y_2Fe_{16.75}Ru_{0.25}$	-	<0.1*	<0.1*
$Y_2Fe_{16.5}Ru_{0.5}$	0.3	0.8	1.4
$Y_2Fe_{16.25}Ru_{0.75}$	0.2	0.4	0.8
$Y_2Fe_{16}Ru$	0.1*	0.2	0.4

*One should keep in mind the standard error of the X-ray dilatometry of ~10^{-4}.

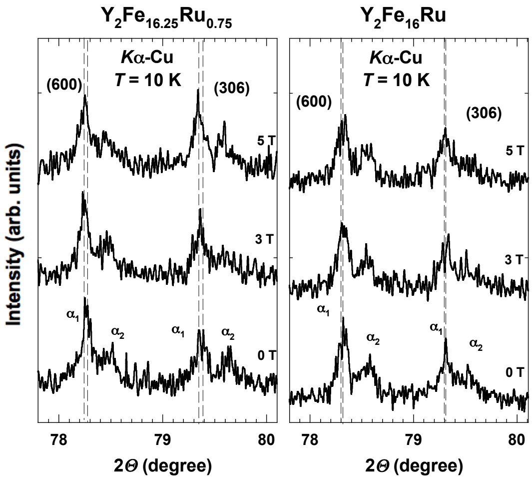

Fig. 5. The (600) and (306) reflection profiles of $Y_2Fe_{16.25}Ru_{0.75}$ (left) and $Y_2Fe_{16}Ru$ (right) powder samples measured in Cu*K*α radiation at 10 K in various fields. The dashed lines indicate the positions of the α_1-(600) and α_1-(306) reflections at 0 and 5 T.

Stronger influence of the second stage of magnetization process on the interatomic distances of the $Y_2Fe_{17-x}Ru_x$ compounds is indicated in Fig. 5, which shows the XRD results obtained for the $Y_2Fe_{16.25}Ru_{0.75}$ and $Y_2Fe_{16}Ru$ compounds at 10 K. A “finite” (in terms of the magnetic field applied) metamagnetic transition in $Y_2Fe_{16.25}Ru_{0.75}$ is manifested in a two-fold volume increase as compared to $Y_2Fe_{16}Ru$ (see Table 1), where the field of 5 T was not sufficient for the development of a nearly-ferromagnetic state. On the other hand, the low magnetostrictive strains in $Y_2Fe_{16}Ru$ qualitatively correspond to the lowest magnetic moment (in general, ω is proportional to M^2) across the $Y_2Fe_{17-x}Ru_x$ series of compounds, showing the low-temperature metamagnetism.

4. Conclusions

The magnetization study of the effect of Ru substitution for Fe on magnetic states of $Y_2Fe_{17-x}Ru_x$ has shown that the collinear ferromagnetic ground state present in the parent Y_2Fe_{17} is destroyed with the increase of Ru content. For the alloys with x > 0.25, the antiferromagnetism is stabilized in the whole temperature range of magnetic order. The $Y_2Fe_{17-x}Ru_x$ compounds possess an easy-plane type of magnetic anisotropy. The field-induced hysteretic transitions at 10 K in $Y_2Fe_{17-x}Ru_x$ (x = 0.5, 0.75) are accompanied by magnetostrictive strains both in the basal plane and along the *c*-axis of the hexagonal Th_2Ni_{17}-structure. The maximum volume change of 0.14% is observed in $Y_2Fe_{16.5}Ru_{0.5}$ at 5 T that is comparable to 0.2% in $Lu_2Fe_{16.5}Ru_{0.5}$ (will be published elsewhere).

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