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Magnetic Anisotropy of $Lu_2Co_{17-x}Si_x$ Single Crystals

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Magnetic anisotropy of Lu₂Co_{17-x}Si_x single crystals grown by the Czochralski method was investigated. The homogeneity range of Si substitution for Co extends up to x = 3.4 in Lu₂Co_{17-x}Si_x solid solutions. The unit cell volume, Curie temperature, and spontaneous magnetic moment decrease monotonously with increasing Si content. Lu₂Co₁₇ has the easy-plane type of magnetic anisotropy in the ground state, which changes into the easy-axis type by two spin-reorientation transitions of the second-order, the easy-plane easy-cone at $T_{\rm SR1} \sim 680$ K and the easy-cone — easy-axis at $T_{\rm SR2} \sim 730$ K. Upon Si substitution, the observed spin-reorientations shift towards the lower temperatures for Lu₂Co_{17-x}Si_x ($T_{\rm SR1} \sim 75$ K and $T_{\rm SR2} \sim 130$ K in Lu₂Co₁₆Si) and vanish for compounds with $1 < x \leq 3.4$, which have the uniaxial type of magnetic anisotropy in the whole temperature range of magnetic order.

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1. Introduction

Co-sublattice in R_2Co_{17} (R = rare earths) intermetallics [1] adopts substitutions by several elements. Substitutions of the 3*d* transition metals [2], as well as Al, Si, and Ga [3–5], for Co in some cases to a sign reversal of the magnetocrystalline anisotropy (MA) of the Co-sublattice from the easy-plane to the easy-axis. Studies of $R_2(Co, M_{17})$ compounds with non-magnetic R (Ce⁴⁺, Lu³⁺ or Y³⁺) and Gd³⁺ (Gd does not contribute to anisotropy) give information on the Co sublattice anisotropy and its changes upon partial substitution by a third M element. Previous investigations with Si were carried out on polycrystalline samples; however, single crystals are strongly desirable for the magnetic anisotropy investigations. The presence of the spontaneous spin-reorientation transitions (SRT) from the easy-plane to the easy-axis was reported in [4, 6, 7] along with the change of the MA type upon Si substitution for compounds with Y and Gd, whereas Ce₂Co_{17-x}Si_x compounds were found to be uniaxial for all Si content. There are some works dedicated to the study of magnetic anisotropy of Lu₂Co₁₇ single

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crystal up to 600 K [8, 9], however, no investigations on the substitution in the Co-sublattice by a non-magnetic element M were performed. Besides, a detailed analysis of available data revealed the fact that at higher temperatures, Lu_2Co_{17} also undergoes SRT [10] from the easy-plane to the easy-axis. But the order of this transition was not clarified yet.

2. Results and discussion

In the present work, the systematic study of the effect of substitution of Si for Co on magnetic anisotropy was performed for $\text{Lu}_2\text{Co}_{17-x}\text{Si}_x$ (x = 0-3.4) single crystals grown by the Czochralski method in a tri-arc furnace. The X-ray powder diffraction analysis confirmed the hexagonal crystal structure of the Th₂Ni₁₇-type [11] for all Lu₂Co_{17-x}Si_x compounds within the homogeneity range (x = 0-3.4). Alloys with $x \ge 3.4$ were found to be multiphase.

The unit-cell volume V of $\text{Lu}_2\text{Co}_{17-x}\text{Si}_x$ decreases monotonously with increasing x (Fig. 1). The lattice parameter a decreases slightly up to x = 2 and then does not depend on x for x > 2, whereas the parameter c is almost constant up to x = 2 and starts to decrease at $x \ge 2$. No changes in the crystal structure type were indicated by the powder X-ray diffraction for compounds with $x \ge 2$.



Fig. 1. Concentration dependences of structural and magnetic properties of $Lu_2Co_{17-x}Si_x$: (a) lattice parameters a, c; (b) unit-cell volume V and c/a ratio; (c) spontaneous magnetic moment M_s (at 5 K) and Curie temperature T_C ; (d) anisotropy constant K_1 (5 K, 300 K).

Figure 2 shows the magnetization curves along the principal axes for single crystals of $Lu_2Co_{17-x}Si_x$ (x = 0, 1) at different temperatures. Both compounds have the easy-plane type of anisotropy in the ground state, which is changed into the easy-axis one upon varying temperature. The type and strength of MA of the

Co sublattice in R_2Co_{17} is a result of competitive contributions of the individual anisotropies (with different signs and temperature dependences) of Co atoms located at different crystallographic sites of the Th_2Ni_{17} -type crystal structure which leads to spin-reorientations. Since the second anisotropy constant $K_2 > 0$, the SRT occurs as two second-order type phase transitions, easy-plane–easy-cone and easy-cone–easy-axis [12]; $T_{SR1} \sim 680$ K and $T_{SR2} \sim 730$ K in Lu₂Co₁₇ (see the inset of Fig. 2b and Fig. 3).



Fig. 2. Magnetization curves along the principal axes of Lu₂Co₁₇ (a,b) and Lu₂Co₁₆Si (c,d) single crystals at different temperatures. The insets in (b,d) present the temperature dependences of the angle Θ between the *c* axis and the easy-magnetization axis determined from the magnetization isotherms.

The easy-plane anisotropy of R_2Co_{17} with non-magnetic R is determined by Co atoms on 6g and 12k sites [4]. Si primarily substitutes Co in these positions and therefore weakens the easy-plane contribution to the total anisotropy. It leads to a different temperature of compensation of the individual anisotropies from different Co sites in Lu₂Co₁₆Si compound. Thus the spin-reorientation transitions shift toward the lower temperatures, $T_{\rm SR1} \sim 75$ K and $T_{\rm SR2} \sim 130$ K. With a further increasing of Si content, the easy-axis contribution from 4f and 12j sites starts to dominate, resulting in the change of the sign of K_1 (Fig. 1d) and in disappearance of the SRTs for Lu₂Co_{17-x}Si_x with x > 1. The temperature dependences of K_1 and K_2 for Lu₂Co_{17-x}Si_x are presented in Fig. 3.



Fig. 3. Temperature dependences of anisotropy constants K_1 and K_2 of Lu₂Co_{17-x}Si_x.

The results show that the substitution of Si for Co in $Lu_2Co_{17-x}Si_x$ leads to the onset of the uniaxial type of anisotropy in compounds with x > 1 with a subsequent decrease of anisotropy due to a dilution effect for compounds with x > 2.

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