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High-field magnetization study of a HoFe₆Al₆ single crystal

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ABSTRACT

Magnetization measurements have been performed in static (up to 14 T) and pulsed (up to 60 T) magnetic fields on a HoFe₆Al₆ single crystal with a tetragonal crystal structure of ThMn₁₂ type. HoFe₆Al₆ is a ferrimagnet with exact compensation of the Ho and Fe sublattices at low temperatures, the spontaneous magnetic moment is $M_s = 0$ at 4.2 K. At elevated temperatures, M_s passes through a wide maximum (4.2 µ_B at 200 K) and vanishes at the Curie temperature, $T_C = 315$ K. The compound displays a high magnetic anisotropy of the easy-plane type, a noticeable anisotropy exists also within the easy plane with the [110] axis as the easy magnetization direction. HoFe₆Al₆ exhibits two field-induced magnetic phase transitions along the easy [110] axis and two more along the [100] axis, the hard direction in the basal plane. The critical fields of the lower- and higher-field transitions display qualitatively different temperature dependencies. A *T*–*H* magnetic phase diagram has been constructed for the [100] and [110] directions. The Ho–Fe inter-sublattice exchange constant has been determined to be $n_{HoFe} = 3.8$ T/µ_B.

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1. Introduction

Binary intermetallic compounds RT_{12} (*R* is a rare earth element or Y, T is a late 3d metal) having the tetragonal crystal structure of Th Mn_{12} type exist only with T = Mn, nevertheless, the structure can be stabilized by a third element. Compounds $R(T,M)_{12}$ (M is a stabilizing element) are a wide group of materials (see Ref. [1] for review). The Fe content x in the RFe_xM_{12-x} quasibinaries may vary in a wide interval. Compounds of this group with a high Fe content, RFe₁₁Ti, RFe_{10.5}V_{1.5}, RFe₁₀Si₂ and RFe₁₀Mo₂, are considered as promising materials for permanent magnets due to their good hard magnetic properties [2–6]. On the other hand, the combination of the localized 4f and itinerant 3d magnetism leads to complex magnetism and makes these materials interesting from a more fundamental viewpoint. In particular, RFe_xAl_{12-x} systems are characterized by competitive exchange and anisotropic interactions within their homogeneity range, $4 \le x \le 6$, and therefore deserve special attention.

The main structural feature of RFe_xAl_{12-x} compounds with respect to their magnetic properties is the gradual filling of the 8j

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concentration order at temperatures above 300 K [34–37]. The compounds with R–Tb, Ho and Er are collinear ferrimagnets [38]. The magnetic moments of the vast majority of RFe_xAI_{12-x} compounds lie in the basal plane of the tetragonal lattice due to the easy-plane contributions provided by the R and Fe sublattices

site by Fe atoms (Fig. 1). In Al-rich *R*Fe₄Al₈, the Fe atoms reside exclusively on the 8*f* site, while the Al atoms can be found in the 8*j*

and 8*i* positions [7–12]. For higher Fe concentrations, Fe atoms

begin to occupy the 8*j* site that they share with Al as was found for

*R*Fe₅Al₇ and *R*Fe₆Al₆ [8,13–20]. As a consequence, the type of ex-

change interactions within the Fe sublattice strongly depends on

the Fe content. For non-magnetic *R* elements, the Fe sublattice in

*R*Fe₄Al₈ was found to be antiferromagnetically ordered [11,21–24].

In Refs. [9,25-28] cycloidal and amplitude-modulated structures were also reported. Interestingly, in RFe_4Al_8 compounds with

magnetic rare-earth elements the *R*–Fe inter-sublattice exchange

interaction is so weak that the R and Fe sublattices order inde-

pendently at different temperatures, below 80 K and above 150 K,

respectively [7,9,29–32]. The exchange interactions in RFe₅Al₇ are

strengthened due to a higher Fe content, and a tendency toward a

ferromagnetic order is observed within the Fe sublattice

[13,14,18,33]. The rare-earth and iron sublattices order at the same

temperature above 200 K. Collinear ferrimagnetic structures were

found by powder neutron diffraction for the compounds with

R–Tb, Dy, Ho, Er and Tm [18]. *R*Fe₆Al₆ systems with the highest Fe









Fig. 1. Crystal structure of RFe_xAl_{12-x} ($4 \le x \le 6$, ThMn₁₂ type).

[9,10,25,27,28,37–40]. The only exception reported so far is TmFe₅Al₇ whose magnetic moments are aligned along the c axis at low temperatures due to the uniaxial Tm magnetic anisotropy, while at higher temperatures the easy-plane Fe anisotropy dominates [41]. Strong magnetic hysteresis was reported for a number of RFe₅Al₇ and RFe₆Al₆ compounds from measurements on polycrystalline samples, which suggested strong magnetic anisotropy [13,14,35]. A systematic investigation of the whole series of ferrimagnetically ordered RFe₅Al₇ (R-Gd-Tm) on single crystals established a strong single-ion anisotropy [40–51]. It is important to note that in RFe₅Al₇ with R–Tb, Dy, Ho and Er at low temperatures the rare-earth sublattice dominates the total magnetization, whereas at higher temperatures this situation changes in favor of the Fe sublattice. For this reason, RFe₅Al₇ display a compensation point, T_{comp}, that varies from 93 K for DyFe₅Al₇ to 31 K for ErFe₅Al₇. Since the effects of magnetic anisotropy are most visible in the vicinity of *T*_{comp} [52], in this temperature range the magnetization of RFe₅Al₇ exhibits complex behavior as a function of magnetic field manifested by field-induced phase transitions. The transitions are observed along the easy magnetization direction and reflect simultaneous rotations of the R and Fe magnetic moments with the transition field passing through zero at the compensation point.

In the present work, we study magnetic properties of the HoFe₆Al₆ compound. For this system, at low temperatures the magnetic moments of the rare-earth and iron sublattices should be close in absolute value, ~10 μ_B , so that the compensation point is shifted to around zero as compared to $T_{\rm comp} = 65$ K for HoFe₅Al₇ [46,50]. HoFe₆Al₆ is expected to be a strongly anisotropic ferrimagnet with a complex behavior in applied magnetic field. The goal of this work is to study the inter-sublattice exchange interactions and magnetic anisotropy of HoFe₆Al₆ using static and pulsed magnetic fields.

2. Experimental details

A single crystal of HoFe₆Al₆ was grown by a modified Czochralski method in a tri-arc furnace from a stoichiometric mixture of the pure elements (99.9% Ho, 99.98% Fe and 99.999% Al). Standard powder X-ray diffraction analysis performed on a part of the single crystal crushed into powder was used to check the crystal structure and to determine the lattice parameters. The X-ray diffraction patterns were refined by means of Rietveld analysis using the Fullprof/Winplotr software [53]. The analysis confirmed a single-phase state with the tetragonal ThMn₁₂-type structure. The lattice parameters, a = 864.2 pm and c = 503.9 pm, are in good agreement with literature [1]. Back-scattered Laue patterns were used to confirm the monocrystalline state and to orient the crystals to cut the samples for magnetization measurements.

The field dependence of the magnetization up to 14 T was measured at different temperatures between 2 and 350 K for fields

applied along the principal crystallographic directions [100], [110] and [001] of a 30 mg sample using a PPMS magnetometer (Quantum Design). PPMS was also used for specific-heat measurements by the relaxation method between 2 and 350 K in zero magnetic field.

High-field magnetization curves were measured along the main axes between 2 and 100 K in pulsed magnetic fields up to 60 T (pulse duration 20 ms). The magnetization was measured by the induction method using a coaxial pick-up coil system. A detailed description of the set-up is given in Ref. [54]. Absolute values of the magnetization were calibrated using data from measurements in static magnetic field.

3. Results and discussion

The upper panel in Fig. 2 presents magnetization curves measured along the principal crystallographic directions of the HoFe₆Al₆ single crystal at 4.2 K (all static-field magnetization curves were obtained in descending magnetic field). The compound displays a zero spontaneous magnetic moment, $M_{\rm s} = 0$. By contrast, at higher temperatures a non-zero spontaneous moment develops (lower panel in Figs. 2 and 3). As outlined in the Introduction, it indicates that at low temperatures a ferrimagnet HoFe₆Al₆ displays a compensation of the Ho and Fe sublattice magnetization. Assuming that at 4.2 K the Ho magnetic moment is equal to its free ion value, $M_{\rm Ho} = 10 \ \mu_{\rm B}$, and the compound is a collinear ferrimagnet, the total moment of the Fe sublattice should be 10 μ_B as well. This yields $M_{\rm Fe} \approx 1.7 \mu_B$ per Fe atom which is close to 1.6 μ_B found for RFe₅Al₇ [39–51]. It follows from the magnetization curves at 40 and 80 K that the magnetic moments of HoFe₆Al₆ lie in the basal plane of the tetragonal lattice. There is no projection of the spontaneous magnetic moment onto the [001]



Fig. 2. Magnetization curves along the main crystallographic axes of HoFe₆Al₆ at low temperatures.

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