ARTICLE IN PRESS

Physica B xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Physica B

journal homepage: www.elsevier.com/locate/physb

Magnetic characteristics of polymorphic single crystal compounds $DyIr_2Si_2$

Kiyoharu Uchima[a,*], Toru Shigeoka[b,*], Yoshiya Uwatoko[c]

[a] *General Education, Okinawa Christian Junior College, Nishihara, Okinawa 903-0207, Japan*
[b] *Graduate School of Science and Engineering, Yamaguchi University, Yamaguchi 753-8512, Japan*
[c] *Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan*

ARTICLE INFO

Keywords:
$DyIr_2Si_2$
Polymorphic compound
Antiferromagnetism
Metamagnetism
Magnetic anisotropy
Crystal field effects

ABSTRACT

We have confirmed that the tetragonal ternary compound $DyIr_2Si_2$ shows polymorphism; the $ThCr_2Si_2$-type structure as a low temperature phase (I-phase) and the $CaBe_2Ge_2$-type one as a high temperature phase (P-phase) exist. A comparative study on magnetic characteristics of the morphs was performed on the I- and P-phase single crystals in order to elucidate how magnetic properties are influenced by crystallographic symmetry. The magnetic behavior changes drastically depending on the structure. The $DyIr_2Si_2$(I) shows an antiferromagnetic ordering below T_N = 30 K, additional magnetic transitions of T_1 = 17 K and T_2 = 10 K, and a strong uniaxial magnetic anisotropy with the easy [001] direction. The [001] magnetization shows four metamagnetic transitions at low temperatures. On the other hand, the $DyIr_2Si_2$(P) has comparatively low ordering temperature of T_{N1} = 9.4 K and an additional transition temperature of T_{N2} = 3.0 K, and exhibits an easy-plane magnetic anisotropy with the easy [110] direction. Two metamagnetic transitions appear in the basal plane magnetization processes. In both the morphs, the χ-T behavior suggests the existence of component-separated magnetic transitions. The *ab*-component of magnetic moments orders at the higher transition temperature T_{N1} for the P-phase compound, which is contrast to the I-phase behavior; the *c*-component orders firstly at T_N. The crystalline electric field (CEF) analysis was made, and the difference in magnetic behaviors between both the morphs is explained by the CEF effects.

1. Introduction

The tetragonal ternary compounds RIr_2Si_2 (R = rare earth = rare earth) exhibit polymorphism; they have two different crystallographic structures: the $ThCr_2Si_2$-type structure ($I4/mmm$) of low temperature phase and the $CaBe_2Ge_2$-type one ($P4/nmm$) of high temperature phase [1], which is referred to as the I-phase and P-phase (I and P are quoted from the symbol of corresponding space group.), respectively. The crystal structure of the I-phase has centrosymmetry, while one of the P-phase is non-centrosymmetry. So this RIr_2Si_2 family is suitable to study how physical properties are influenced by crystallographic symmetry without changing chemical composition. Bazela presented the correlation between the crystal structure and magnetic ordering, and asserted a strong dependence of magnetic ordering on the a/c ratio [2]. The comparative studies with magnetic behaviors of two polymorphs on single crystal compounds are interesting. Comparative studies with physical properties of polymorphs have been performed on some compounds of this family. The high temperature phase (P-phase) compounds of YIr_2Si_2 and $LaIr_2Si_2$ become superconducting at 2.52 K and 1.24 K, respectively, while the low-temperature ones (I-phase) are normal down to 1 K [3]. Both $CeIr_2Si_2$ polymorphs remain paramagnetic. The I-phase compound behaves as a Fermi-liquid, whereas the P-phase one exhibits non-Fermi-liquid features [4]. In $PrIr_2Si_2$, the P-phase compound remains paramagnetic down to 2 K whereas the I-phase one exhibits an antiferromagnetic ordering below 45.5 K. The different types of magnetic anisotropy, an easy axis anisotropy in the I-phase compound and an easy plane one in the P-phase one, has been reported [5]. Magnetism in $NdIr_2Si_2$ has been studied only for the I-phase compound. It shows a collinear antiferromagnetic ordering below 32.3 K with Nd magnetic moments along the c-axis [6]. For RIr_2Si_2 (R = heavy rare earth = heavy rare earth) series, there are a few reports on I-phase polycrystalline compounds; $TbIr_2Si_2$, $DyIr_2Si_2$ and $ErIr_2Si_2$ order antiferromagnetically below 80 K (or 75 K), 40 K and 10 K, respectively [7–10]. We have already reported the magnetic properties of $TbIr_2Si_2$ single crystals; The $TbIr_2Si_2$(I) shows an antiferromagnetic ordering below T_N = 80 K, a strong uniaxial magnetic anisotropy with the easy [001] direction, and isotropic magnetic behavior in the basal plane. On the other hand, the $TbIr_2Si_2$ (P) has the comparatively low ordering temperature of T_{N1} = 11.5 K and an additional transition temperature of T_{N2} = 5 K, and

---

[*] Corresponding authors.
E-mail addresses: uchima@ocjc.ac.jp (K. Uchima), shigeoka@yamaguchi-u.ac.jp (T. Shigeoka).

http://dx.doi.org/10.1016/j.physb.2017.09.038
Received 27 June 2017; Received in revised form 12 September 2017; Accepted 13 September 2017

Please cite this article as: Uchima, K., Physica B (2017), http://dx.doi.org/10.1016/j.physb.2017.09.038

exhibits an easy-plane magnetic anisotropy with the easy [100] direction. The crystalline electric field effects estimated in the non-Kramers $TbIr_2Si_2$ compound well explain the magnetic anisotropy [11]. The magnetic behavior of Kramers $DyIr_2Si_2$ is also interesting.

We succeeded to grow both $DyIr_2Si_2$(I) and (P) single crystals, and performed magnetic measurements on the single crystals. In this paper, we present comparative magnetic behaviors and the crystalline electric field (CEF) analyses on both the morphs.

2. Experimental procedure

Single-crystals $DyIr_2Si_2$ were grown by the Czochralski technique using a tri-arc furnace; that is, polycrystalline samples were prepared by arc-melting a stoichiometric mixture of the pure elements (Dy: purity of 3N, Ir: 3N, and Si: 5N) in an argon atmosphere. Subsequently, a single crystal was grown by the pulling-up method in the tri-arc furnace. The grown crystal was confirmed to be the P-phase and the single phase nature from analysis of X-ray powder diffraction pattern. A single crystal of the I-phase was obtained by annealing a part of the grown single crystal at 800 °C for 7 days. The crystal structure and the single phase nature of the annealed crystal were also confirmed by X-ray powder diffraction. The quality of the single crystals was checked, and crystallographic orientations were determined by the back Laue method. The crystal was fixed on a thin plastic plate so that the desired direction is perpendicular to the plate within an experimental angle accuracy of one degree. It was subjected to magnetic measurements. The magnetic susceptibility (which is determined from M/B under $B = 0.1$ or 0.01 T where magnetization curves are linear.) and low-field magnetization below 5 T and/or 7 T were measured using a MPMS and/or PPMS (Quantum Design). Pulsed high-field magnetization measurements were performed up to 57 T by a pick-up coil method using a non-destructive long-pulse magnet installed at the International MegaGauss Science Laboratory in the Institute for Solid State Physics, the University of Tokyo.

3. Results and discussion

3.1. Lattice parameter

X-ray diffraction measurements were done to determine to which phase crystals belong. Figs. 1 and 2 show powder diffraction patterns on $DyIr_2Si_2$(I) and (P) compounds, respectively at a room temperature. All reflections in Figs. 1 and 2 could be indexed on the tetragonal $ThCr_2Si_2$- and the $CaBe_2Ge_2$-type structures, respectively, confirming the single phase natures. Then we were able to obtain crystallographic data of both the compounds by least square analysis to the powder pattern, which are shown in the Table 1. The lattice parameters of the I-phase compound are almost in agreement with the previous report [10]. The c/a ratio of the I-phase compound is 1.02 times as large as that of the P-phase one; The I-phase compound, low temperature phase one, elongates along the c-axis comparing with the P-phase one, high temperature phase. Bazela presented the correlation between the crystal structure and magnetic ordering, and asserted a strong dependence of magnetic ordering on the c/a ratio [2]. We can expect that this difference make difference of magnetic anisotropy between two morphs, and change magnetic ordering.

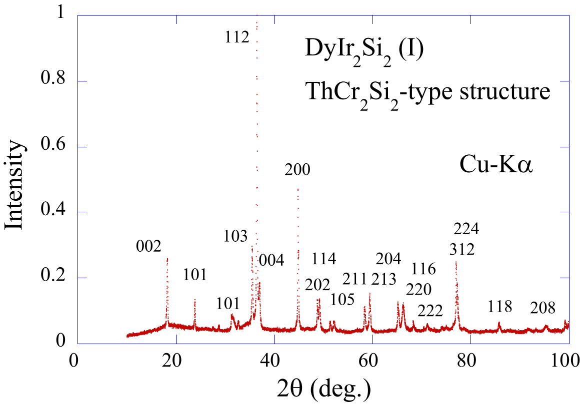

Fig. 1. X-ray (Cu-Kα) diffraction pattern on a $DyIr_2Si_2$(I) powder compound at a room temperature.

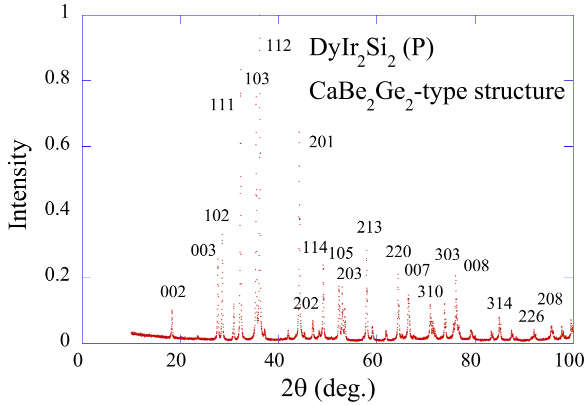

Fig. 2. X-ray diffraction pattern on a $DyIr_2Si_2$(P) powder compound at a room temperature.

Table 1
Crystallographic data of both the $DyIr_2Si_2$ morphs.

$DyIr_2Si_2$	a (nm)	c (nm)	c/a
I	0.405	0.975	2.41
P	0.410	0.967	2.36

3.2. Magnetic susceptibility

The temperature dependences of magnetic susceptibilities along the main symmetry axes in the tetragonal cell for the I- and P-phase compounds are shown in Figs. 3 and 4, respectively. A precise magnetic anisotropy between the c-axis and directions in the basal plane is evidenced from the figures while an anisotropy within directions in the basal plane is very small. In Fig. 3 (the I-phase compound), the [001] susceptibility shows three anomalies at $T_N = 30$ K, $T_1 = 17$ K and $T_2 =$

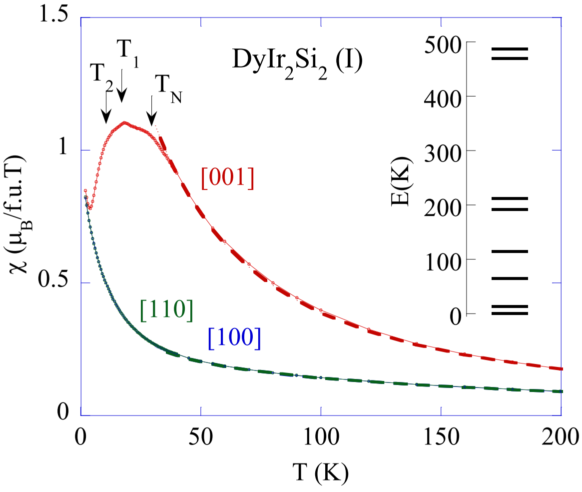

Fig. 3. Temperature dependences of magnetic susceptibilities along the main symmetry axes of a tetragonal cell on a $DyIr_2Si_2$(I) single crystal. Broken lines show calculated values using the CEF parameters. The inset is the level scheme deduced from CEF analysis.