ELSEVIER

Journal of Alloys and Compounds 386 (2005) 82–86

Journal of ALLOYS AND COMPOUNDS

www.elsevier.com/locate/jallcom

Electronic structure and magnetic examination of $ScMn_2$ single crystal

M. Kulpa [a,*], E. Talik [a], A. Winiarski [a], T. Mydlarz [b], A. Gilewski [b], J. Kusz [a,c], H. Böhm [c], W. Suski [b]

[a] *Institute of Physics, University of Silesia, Uniwersytecka 4, 40-007 Katowice, Poland*
[b] *International Laboratory of High Fields and Low Temperatures, Gajowicka 95, 53-529 Wroclaw, Poland*
[c] *Institut für Geowissenschaften der Universität, 55099 Mainz, Germany*

Received 10 April 2004; received in revised form 27 May 2004; accepted 27 May 2004

Abstract

A $ScMn_2$ single crystal was obtained by the Czochralski method from a levitated melt. The electrical resistivity exhibits a negative curvature at higher temperatures. The magnetization versus magnetic field up to 330 kOe was measured at 4.2 K. XPS investigations revealed a splitting of the valence band due to spin fluctuations. This observed splitting of the Mn 3d states proves that the theoretical model of the electronic structure given by Yamada and Shimizu is correct. Lattice parameters were also measured in temperature range 8–300 K.

Keywords: Rare earth compounds; Transition metal compounds; Single crystal; Magnetic properties; Electronic structure; Electrical resistivity

1. Introduction

The RMn_2 intermetallic compounds are the subject of investigations for many years because of the very interesting Mn magnetic moment instability connected with a critical nearest neighbour Mn–Mn distance $d_c = 2.66$ Å. For the compounds with a distance greater than the critical one a localized magnetic moment on the Mn sites is observed (even 2.7 μ_B for YMn_2 [1]). For the RMn_2 compounds with a distance smaller than the critical one, no magnetic moment on the Mn sites is observed, as in the case of the $ErMn_2$ compound [2]. For the compounds with a distance close to the critical, only 1/4 of the Mn atoms are magnetic, which happens in the case of $DyMn_2$ [3] and $HoMn_2$ [4]. The remaining 3/4 of the Mn atoms in $DyMn_2$ and $HoMn_2$ have $\mu_{Mn} = 0$.

RMn_2 compounds crystallize in the cubic C15 Laves phase type of structure for R = Y, Gd, Tb, Dy, Ho and in the hexagonal C14 Laves phase type of structure for R = Sc, Pr, Nd, Sm, Ho, Er, Tm, Yb, Lu. Under high pressure (8 GPa) it is possible to obtain RMn_2 compounds with C14 type of structure for R = Y, Gd, Tb, Dy [5]. Because, the Sc atoms are small in comparison with the R atoms, for $ScMn_2$ the Mn–Mn distance is also the smallest (2.52 Å) in the RMn_2 series. The model of a critical distance predicts that the Mn atoms in $ScMn_2$ do not possess a localized magnetic moment. It was confirmed by NMR measurements that the Mn atoms, in fact, do not carry a magnetic moment [6]. For the $ScMn_2$ compound the bulk modulus was obtained, $K_0 = 80(4)$ GPa [7].

For the YMn_2 compound (C15) the Mn atoms possess localized magnetic moments below the ordering temperature and the magnetic structure is helimagnetic [8]. The effect of replacing Y by Sc is that $Y_{1-x}Sc_xMn_2$ is a paramagnet [9] because of the decrease of the Mn–Mn distance.

2. Experimental

The crystal was grown by the Czochralski method from a levitated melt, using high purity starting materials. The quality of the $ScMn_2$ crystal was checked by X-ray diffraction, using a Siemens D5000 diffractometer with Cu Kα radiation and Berg–Barrett topography using Fe Kα radiation. In Fig. 1, we show a Berg–Barrett topography of a sample. The obtained crystal was without mosaic structure with some irregularities on the surface during the growth process.

Small crystals of $ScMn_2$ suitable for X-ray analysis were isolated by mechanical fragmentation and checked by the

* Corresponding author. Tel.: +48-32-3591740; fax: +48-32-2588431.
E-mail address: mkulpa@us.edu.pl (M. Kulpa).

0925-8388/$ – see front matter © 2004 Elsevier B.V. All rights reserved.
doi:10.1016/j.jallcom.2004.05.075

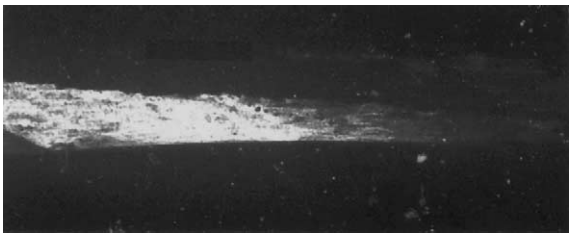

Fig. 1. X-ray Berg–Barrett topography of a $ScMn_2$ single crystal.

X-ray Laue methods. A fragment of a single crystal of the approximate dimension 0.3 mm × 0.3 mm × 0.3 mm was studied using a multilayer monochromator (Osmic) with Cu Kα radiation (Schneider rotating anode) and a four circle Huber diffractometer with a 250 mm χ-circle, controlled by a PC computer with the STOE STADI 4 program system and equipped with a two-stage closed-cycle helium cooling device (CTI-Cryogenics) [10]. The study was performed between 8 K and room temperature. The temperature was controlled within 0.1 K. The refinement of the cell parameters was carried out by measuring 46 reflections with high 2θ values and their Friedel pairs at both sides of the primary beam. An ω-scan was carried out at + and -2θ. The centre of gravity was determined for both scans. The observed 2θ value was given by the difference of the two-ω-scan centres [11]. These results were of zero point errors, absorption effects and systematic errors resulting from a crystal miscentering.

The electrical resistivity temperature dependence was measured along the growth direction [1 3 3] by a conventional method, using a 100 mA current in the temperature range 4.2–350 K starting from 350 K.

The magnetization curves were measured along the growth direction [1 3 3] in static and pulsed magnetic fields up to 330 kOe and at 4.2 K at the International Laboratory of High Field and Low Temperatures in Wrocław. The maximal value of the static magnetic field was 150 kOe.

The XPS electronic structure was measured with monochromatized Al Kα radiation at room temperature, using a Physical Electronics PHI 5700/660 Photoelectron Spectrometer. The spectra of the photoelectrons were analysed by a hemispherical mirror analyser with an energy resolution of 0.3 eV. The spectra were obtained on a single crystal, broken under high vacuum to avoid oxygen and carbon contaminations.

3. Results

3.1. Lattice parameters

The temperature dependence of the lattice parameters was measured for the *a* and *c* parameters and also for the volume (Fig. 2a–c). For the temperature region 150–300 K the thermal expansion coefficients are $\alpha_a = 8.26(8) \times 10^{-5}\,K^{-1}$ for the *a* direction and $\alpha_c = 1.20(8) \times 10^{-4}\,K^{-1}$ for the *c* direction and $\alpha_v = 8.58(6) \times 10^{-4}\,K^{-1}$ for the volume. High values of the thermal expansion coefficients in both the *a* and *c* directions exist because of the existence of spin fluctuations in $ScMn_2$. The amplitude of the spin fluctuations is increasing with temperature. The values of the lattice parameters at room temperature are $a = 5.043(2)$ Å and $c = 8.271(4)$ Å. The solid lines are calculated from the Grüneisen–Debye theory. Different Debye temperatures have been determined from the best fit of the experimental data (lattice parameters versus temperature) for different directions: 320 K for *a* direction, 400 K for *c* direction and 360 K for the volume expansion. As predicted by the model of a critical distance there are not any magneto-volume anomalies in the thermal expansion of $ScMn_2$. Such anomalies are typical for RMn_2 compounds with a localized magnetic moment on the Mn sites (e.g. $GdMn_2$) and they do not appear for RMn_2 compounds with an itinerant Mn moment (e.g. $LuMn_2$) [12].

3.2. Magnetic properties

In Fig. 3, the relation between magnetization and magnetic field measured at 4.2 K is shown. The data for static fields and for pulsed fields form one curve. The values of the magnetization are small (0.14 μ_B/f.u. at 350 kOe), because $ScMn_2$ is a Pauli paramagnet. No hysteresis loop was observed. The magnetization curve is slightly bent below 50 kOe and forms a straight line in the range 50–350 kOe. For $ScMn_2$ at 30 kOe, the magnetization has the value 0.036 μ_B/f.u. and for C14 YMn_2 0.08 μ_B/f.u. [13] and only 0.006 μ_B/f.u. for C15 YMn_2 [14]. The magnetization versus magnetic field curve measured at 4.2 K for $ScMn_2$ is similar to that measured for C14 YMn_2, because both compounds have non-magnetic Mn atoms. On the contrary, the values of magnetization for C15 YMn_2 are much lower because the Mn atoms posses a localized magnetic moment and they form a canted antiferromagnetic structure (helimagnetic).

3.2.1. Electrical resistivity

The temperature dependence of the electrical resistivity is shown in Fig. 4. The relation $\rho = \rho_0 + AT^3$ ($A = 5.31 \times 10^{-7}\,\mu\Omega m/K^3$, $\rho_0 = 0.25\,\mu\Omega m$) is exactly satisfied up to 70 K. A similar T^3 behaviour of the electrical resistivity was also observed for $DyMn_2$ [15]. The curvature of the resistivity versus temperature curve changes from positive ($T < 150$ K) to negative ($T > 150$ K). The negative curvature is characteristic for spin fluctuations. A very similar temperature dependence of the electrical resistivity, due to spin fluctuations, was observed for the $LuMn_2$ compound [16], although a T^2 relation was satisfied at low temperatures.

3.3. Electronic structure

All the spectra were collected using a fresh surface after breaking a sample under high vacuum (~10^{-10} Torr). In Fig. 5 can be seen that near the Fermi level splitted states are present. The states result from hybrydization between Sc 3d and Mn 3d states. The splitting can be also linked with