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Magnetic properties of new compounds $RuMn_2Sn$ and $RuMn_2Si$

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ABSTRACT

New compounds $RuMn_2Z$ (Z = Si, Sn) have been synthesized. X-ray diffraction measurements have confirmed that $RuMn_2Z$ (Z = Si, Sn) crystallizes in a Heusler-like cubic structure. The lattice parameters of $RuMn_2Si$ and $RuMn_2Sn$ at room temperature are estimated to be 5.8260 Å and 6.2195 Å, respectively. Magnetization measurements have been carried out in fields up to 50 kOe for $RuMn_2Z$ (Z = Si, Sn). Furthermore, the temperature dependence of initial permeability of $RuMn_2Sn$ has been studied. $RuMn_2Sn$ shows ferrimagnetic behavior. The spontaneous magnetic moment at 5 K and the Curie temperature of $RuMn_2Sn$ are found to be of 1.68 μ_B/f.u. and 272.1 K, respectively. $RuMn_2Si$ exhibits spin-glass-like behavior with a freezing temperature estimated to be about 50 K.

1. Introduction

Ferromagnetic shape memory alloys (FSMAs) with the $L2_1$ structure have attracted much attention due to their potential application as smart materials [1,2]. They show a large magnetic-field-induced strain by the rearrangement of twin variants in the martensite phase [3]. Until now, several candidates for FSMAs have been reported. Among them, the stoichiometric Heusler alloy Ni_2MnGa is the most studied system. Ni_2MnGa has the cubic $L2_1$ Heusler structure at room temperature and orders ferromagnetically at a Curie temperature of 365 K. On cooling below a martensitic transition temperature of about 200 K, a superstructure forms [4]. A number of investigations on Ni–Mn–Ga FSMAs have been described in the literature because the Curie temperature and the martensitic transition temperature can be tuned by changing the composition of the constituent elements. Topics relating to Ni–Mn–Ga FSMAs have been reviewed by Entel et al. [5] and Brown et al. [6] in recent articles.

Recently, Liu et al. synthesized a new compound $NiMn_2Ga$ which carries a magnetic moment of about 1.4 μ_B/f.u. at 5 K [7]. The Curie temperature was found to be 588 K [7]. Furthermore, they reported that $NiMn_2Ga$ exhibits a martensitic transition around room temperature with a large hysteresis up to 50 K and a lattice distortion as large as 21.3%. They also observed an excellent two-way shape memory behavior with a strain of 1.7% in a single crystal. Both experimental and theoretical studies made clear that $NiMn_2Ga$ crystallizes in the Hg_2CuTi-type structure (space group $F\bar{4}3m$), which is different from the $L2_1$ structure with $Fm\bar{3}m$ space group [8]. Simultaneously, Barman et al. discussed the martensitic transition, ferrimagnetism and Fermi surface nesting using a first-principles calculation [9,10]. Liu et al. investigated theoretically and experimentally the electronic structures and magnetic properties of $CoMn_2Z$ (Z = Al, Ga, In, Si, Ge, Sn and Sb) compounds [11]. It was predicted that the compounds with Z = Al, Si, Ge, Sn and Sb are half-metallic ferrimagnets. Experimentally, they successfully synthesized the $CoMn_2Z$ (Z = Al, Ga, In, Ge, Sn and Sb) compounds and confirmed that these compounds crystallize in the Hg_2CuTi-type structure instead of the conventional $L2_1$ structure. However, there has been only a little amount of information on the magnetic moments and the exchange interactions for XMn_2Z (X = transition element: Z = s, p element) compounds. In this paper, the structural and magnetic properties of $RuMn_2Z$ (Z = Si, Sn) are examined experimentally to gain deeper insight into the magnetic moments and the exchange interactions of XMn_2Z alloys.

2. Experimental

For the preparation of the specimens, powdered ruthenium (99.9%), manganese (99.99%), silicon (99.9999%) and tin (99.99%) were mixed in the desired proportions and sealed in evacuated silica tubes. For the preparation of $RuMn_2Sn$, the mixture was first heated at 1000 °C for 3 days and then quenched into water. To achieve a high crystalline perfection, the reaction product was pulverized, carefully ground and heated again at 1050 °C for 3 days. The reaction product was then reheated to 1100 °C for 3 days and then quenched into water. For the preparation of $RuMn_2Si$, the mixture of constituents was first heated to 1100 °C for 3 days. Thereafter the reaction product was heated to 1100 °C for 3 days and then quenched into water. The

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observed X-ray diffraction patterns were analyzed using the Rietveld profile refinement program. The temperature dependence of the initial permeability was measured using an ac transformer method in which the primary and secondly coils were wound around a cylindrical sample of diameter ~ 1 mm and length ~ 10 mm. When an ac current of a constant amplitude flows in the primary coil, the voltage induced in the secondary coil is directly proportional to the initial permeability. Magnetization measurements on each sample were made in magnetic fields up to 50 kOe using a superconducting quantum interference device (SQUID) magnetometer.

3. Results and discussion

The Heusler $L2_1$-type structure is comprised of four interpenetrating fcc sublattices with A, B, C and D sites. The A, B, C and D sites are located at (0, 0, 0), (1/4, 1/4, 1/4), (1/2, 1/2, 1/2) and (3/4, 3/4, 3/4), respectively, as shown in Fig. 1. Fig. 2(a) shows the experimental X-ray diffraction pattern at room temperature for a powder sample of $RuMn_2Sn$. All the experimental diffraction lines can be indexed with the cubic structure. The sharp (2 2 0) peak confirms the presence of a single cubic phase. The lattice parameter a of $RuMn_2Sn$ at room temperature was found to be 6.2195 Å. The pattern in Fig. 2(b) was calculated using the $L2_1$ structure and the experimental lattice parameter, with the A and C sites occupied by the Mn atoms, and B and D sites occupied by Ru and Sn atoms, respectively. We call this structure the $L2_1$A-type. By exchanging the Mn atoms at the C sites with the Ru atoms at the B sites, the $L2_1$A-type structure changes into another ordered arrangement with a different superstructure. The prototype of this structure is the Hg_2CuTi compound which has the $F\bar{4}3m$ space group. Fig. 2(c) shows the X-ray powder diffraction pattern calculated by assuming the Hg_2CuTi-type structure. We call this structure the XA-type. The X-ray powder diffraction pattern of Fig. 2(d) was calculated by assuming that Mn atoms and Sn atoms occupy B sites and D sites, respectively, and that the other Mn atoms and Ru randomly occupy the A and C sites. This leads to the $L2_1$ structure though the atomic arrangement is different from the $L2_1$A-type structure. We call this structure the $L2_1$B-type. The intensity I of different lattice diffraction line is proportional to the square of the structure factor, $F(h\,k\,l)^2$ in which h, k and l are the Miller indices. For Heusler alloys, $F(1\,1\,1)$, $F(2\,0\,0)$ and $F(3\,1\,1)$, etc. correspond to the order-dependent superlattice diffraction lines, whilst $F(2\,2\,0)$ is an order independent fundamental diffraction line. Therefore, the different superlattice structures can be distinguished by comparing the intensity ratio

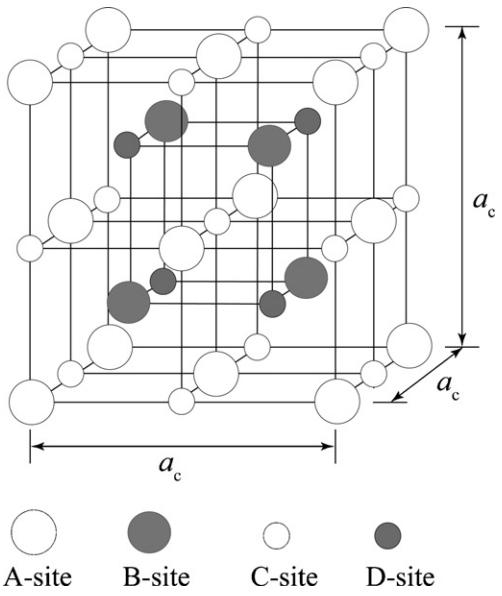

Fig. 1. Crystal structure of the Heusler-type alloy.

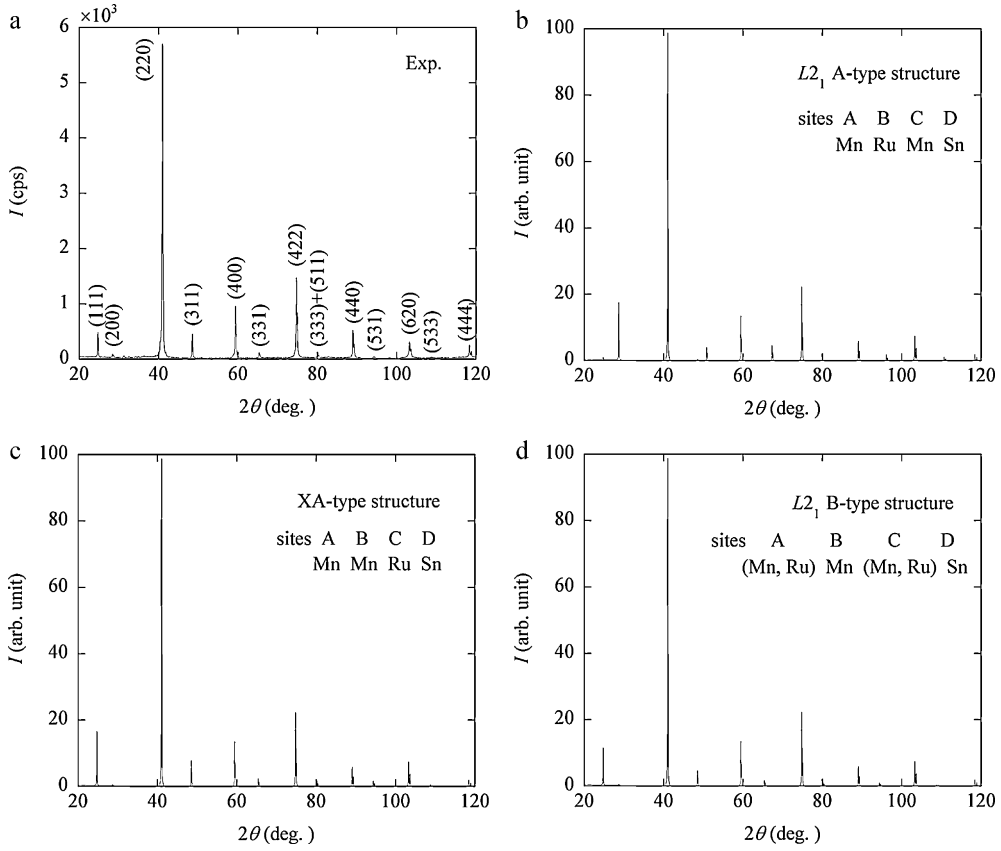

Fig. 2. Observed X-ray powder diffraction pattern at room temperature of $RuMn_2Sn$ (a). Calculated X-ray powder diffraction pattern of $RuMn_2Sn$ for the $L2_1$A-type crystal structure (b), the XA-type crystal structure (c) and the $L2_1$B-type crystal structure (d). The $L2_1$A, XA and $L2_1$B-type crystal structures are explained in the text.