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Magnetic properties of $Co_2Fe(Ga_{1-x}Si_x)$ alloys

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ABSTRACT

Magnetic and crystallographic properties of bulk $Co_2Fe(Ga_{1-x}Si_x)$ alloys with $0 \le x \le 1$ are reported in this work. The alloys with $x=0.75$ and 1.00 exhibit $L2_1$ structure whereas the alloys with $x=0$, 0.25 and 0.50 crystallized in the disordered A2 phase. Unit cell volume of this series of alloys decreased from 189.1 to 178.5 Å^3 as x was increased from 0 to 1.00. All alloy compositions exhibit ferromagnetic behavior with a high Curie temperature (T_C) which showed a systematic variation with x (1089 K, 1075 K, 1059 K, 1019 K and 1015 K for $x=0$, 0.25, 0.5, 0.75 and 1.00, respectively). The saturation magnetization moment M_s for the alloys with $x=0$, 0.25 and 0.50 are 5.05μ_B, 5.23μ_B, 5.49μ_B, respectively, in accordance with the Slater–Pauling rule, but alloys with $x=0.75$ and 1.00 deviated from this rule. The effective moment per magnetic atom (p_c) of the alloys was estimated from the inverse DC magnetic susceptibility data above T_C. A comparison of M_s with p_c reveals the half-metallic character of the alloys.

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1. Introduction

Full Heusler alloys are ternary intermetallic compounds with a chemical formula of X_2YZ, where X and Y are transition metals and Z is a sp-valent element. The stable $L2_1$ structure of these alloys consists of four interpenetrating FCC sub-lattices with Wyckoff positions of (0 0 0), (¼ ¼ ¼), (½ ½ ½) and (¾ ¾ ¾) [1–3]. This class of alloys exhibits a wide variety of magnetic properties like ferromagnetism, ferrimagnetism, antiferromagnetism, Pauli paramagnetism etc. [3]. Prediction of half-metallic behavior first in Ni–Mn–Sb [2] and subsequently in Co_2MnSi [4,5] have intensified the interest in these alloys. Half-metallic ferromagnets are characterized by coexistence of metallic behavior for one spin direction and semiconducting behavior with energy band gap for other spin direction. This novel electronic structure results in 100% spin polarization at the Fermi level. The total spin magnetic moment per formula unit for X_2YZ alloys is given by the Slater–Pauling (S–P) relation,

$$M_t = (Z_t - 24)\mu_B \quad (1)$$

where Z_t is the total number of valence electrons in the unit cell [6].This relationship provides a clue for identifying new half-metallic alloys suited for fabricating spintronic devices. In this context, Co_2YZ and Fe_2YZ based alloys have received a lot of attention because of their high Curie temperature and high magnetic moment as well as theoretically predicted half metallic nature [7–15]. Among these systems, Co_2FeSi alloy has been extensively studied. Reports [9,12] show that the magnetic moment of this alloy is in the range of 5.18–5.97μ_B. On the other hand, the magnetic moment and T_C of Co_2FeGa alloy have been reported to be 5.17μ_B and 1093 K, respectively [11]. Considering the advantage of fourth element substitution in the ternary full Heusler alloys, we report the effect of systematic substitution of Si for Ga in Co_2FeGa alloy on its structural and magnetic properties with a view to develop new alloys for possible spintronic applications.

2. Experimental

$Co_2Fe(Ga_{1-x}Si_x)$ ($0 \le x \le 1$) alloy ingots were prepared by arc melting appropriate amounts of high purity ($\ge$ 99.99%) elements in argon atmosphere. After melting several times, the weight loss in the alloy ingot was found to be less than 2% of the initial weight. The ingots were then taken in sealed fused silica ampoules evacuated to 10^{-3} Pa. The ingots in the sealed ampoules were annealed at 1273 K for 3 days and quenched in ice water. Crystal structure of the alloys was determined with a powder X-ray diffractometer (Seifert, 3003T/T) emitting Mo K_α X-rays ($\lambda=0.07093$ nm). Magnetic properties were measured using a vibrating sample magnetometer (VSM, Lakeshore 7410) equipped with a variable temperature sample holder. The overall composition of the alloys was verified using an energy dispersive spectrometer (EDS, Oxford) attached to a scanning electron microscope (SEM, Leo 1430 VP) and was found to be within 1% of the nominal composition.

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3. Results and discussion

Room temperature X-ray diffraction (XRD) patterns of $Co_2Fe(Ga_{1-x}Si_x)$ alloys are shown in Fig. 1. A fully ordered Heusler (X_2YZ) alloy with $L2_1$ structure (Fig. 2) gives Bragg reflections with non-zero structure factor when all indices are either even or odd [16,17]. The structure factor of the first three reflections are given by the relations,

$$F(111) = 4(f_y - f_z), \quad F(200) = 4[2f_x - (f_y + f_z)], \quad F(220) = 4[2f_x + f_y + f_z] \tag{2}$$

where f_x, f_y and f_z are average scattering amplitudes for respective sub-lattices. The alloys with $x=0.75$ and $x=1.00$ crystallized in $L2_1$ structure as revealed by the presence of the super-lattice peaks (111) and (200). But these peaks were absent in the XRD patterns of the alloys with high Ga concentration i.e., for $x=0$, 0.25 and 0.50. Though the absence of these super-lattice peaks suggests a disordered A2 structure, it has to be noted that the nearly equal scattering factors of Co, Fe and Ga, could also result in such XRD patterns devoid of these super-lattice peaks [18]. Similar structural features have also been reported for this alloy system by Umetsu et al. [15]. The experimental XRD patterns were refined by the Rietveld method using FullProf 2.00 software. Rietveld refinement plot for Co_2FeSi (i.e., $x=1$) alloy is shown in Fig. 3. As x is increased, the peaks shift towards higher 2θ values. Fig. 4 shows the composition dependence of the lattice constant a for this alloy system. A continuous contraction of lattice occurs with an increase

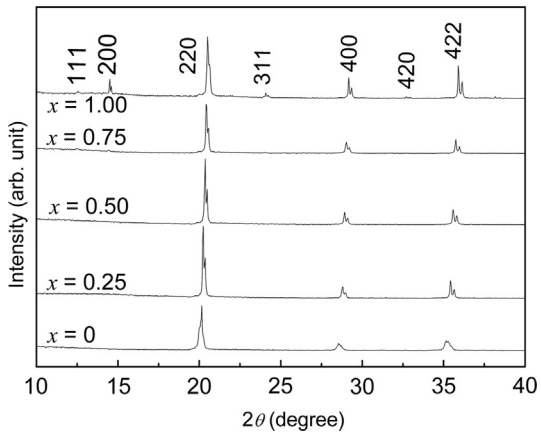

Fig. 1. Room temperature XRD patterns of $Co_2Fe(Ga_{1-x}Si_x)$ alloys with $0 \le x \le 1$.

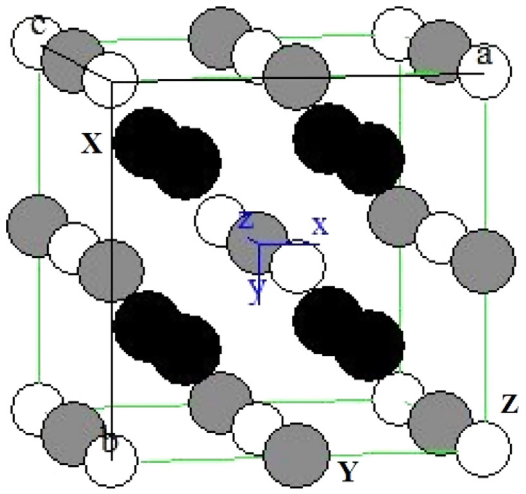

Fig. 2. Arrangement of atoms in an $L2_1$ unit cell with X atoms at 8c (¼ ¼ ¼ and ¾ ¾ ¾), Y atoms at 4b (½ ½ ½) and Z atoms at 4a (0 0 0) Wyckoff positions.

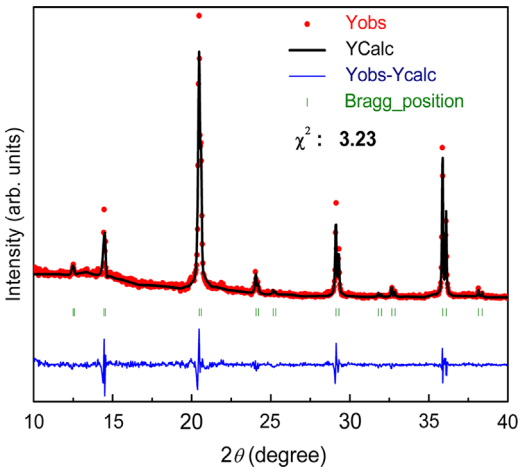

Fig. 3. Rietveld refinement plot of XRD data of Co_2FeSi alloy.

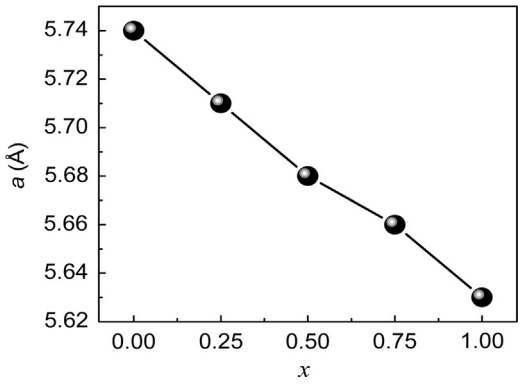

Fig. 4. Variation of lattice constant as a function of Si (x) substitution in $Co_2Fe(Ga_{1-x}Si_x)$ alloys.

in x because the atomic radius of Si (=1.46 Å) is smaller than that of Ga (=1.52 Å). The lattice constant of the end members of this alloy system i.e. Co_2FeGa and Co_2FeSi are found to be 5.74 Å and 5.63 Å respectively, which is in agreement with earlier experimental observations [11,12,15].

Room temperature initial magnetization (M–H) curves obtained for all the alloy compositions are shown in Fig. 5. Saturation magnetization (M_s) for the alloys with $x=0$, 0.25, 0.50, 0.75 and 1.00 are 5.05μ_B, 5.23μ_B, 5.49μ_B, 5.44μ_B and 5.42μ_B, respectively. So the alloys with $x=0$, 0.25 and 0.50 follow the S–P rule i.e. Eq. (1). However M_s for the alloys with $x=0.75$ and $x=1.00$ are lower than the value predicted by the S–P rule. The theoretical and experimental M_s for the alloys are shown in Fig. 6. M_s of the $x=1$ alloy is less than the value of 6.0μ_B predicted by the S–P rule as pointed out in earlier reports (e.g., 5.18μ_B [9], 5.73μ_B [13], 5.87μ_B [15], 5.91μ_B [7] and 5.97μ_B [12]). The large diversity in experimentally obtained results can be attributed to the strong dependency of total magnetic moment on the degree of disorder and the change in chemical composition of Co_2FeSi alloys. Chemical composition of our samples has been verified by EDS studies to be within 1% of the nominal composition. Moreover the presence of (1 1 1) and (2 0 0) super-lattice peaks in its XRD pattern establishes $L2_1$ order in this alloy. So, the probable cause for the lower (i.e., $<6.0\mu_B$) M_s value obtained for our sample could be the presence of a small amount of DO_3 type disorder due to swapping of some Co and Fe atoms. The nearly equal scattering factors of Co and Fe makes this disorder hard to detect from XRD studies [18]. Due to this disorder, magnetic moment of Fe atom on the 8c site (*cf.* Fig. 2) decreases from 3.16μ_B to 2.45μ_B, which is only partly compensated by the moderate

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