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ABSTRACT

We performed combined theoretical and experimental studies of electronic, structural, and magnetic properties of Fe$_2$MnSn Heusler alloy. The density functional theory calculation shows that the Fe$_2$MnSn alloy has two energetically close phases, namely, hexagonal D0$_{19}$ and cubic L2$_1$, which agrees well with the experimental results. Both the hexagonal and cubic phases are ferromagnetic, with large magnetization values of about 6 $\mu_B$/f.u. and high Curie temperature above room temperature. The hexagonal phase shows high magnetic anisotropy of 5.1 Merg/cm$^3$. These findings may be interesting for magnetic applications where large saturation magnetization, high Curie temperature, and high magnetic anisotropy are desired.

I. INTRODUCTION

Heusler compounds have been known for over a century, but recently they have attracted renewed interest for possible applications in spin-based electronics. Many of these materials have been reported to exhibit high spin polarization or half-metallicity. Most of the reported Heusler alloys have crystallized in cubic structure and are classified into four groups, namely, full-Heusler, half-Heusler, inverse-Heusler, and quaternary-Heusler. Recently, some non-cubic (hexagonal and tetragonal) Heusler materials with high spin-polarization have been also found. These materials are investigated as prospective materials for novel devices which employ spin-transfer torque (STT) phenomena such as STT magnetic random access memories (MRAMs).

Heusler materials are interesting because of their high Curie temperature and the magnetic properties that can be tuned with elemental composition to fit specific applications. The non-cubic Heusler compounds are of special importance because they may exhibit large magnetocrystalline anisotropy (MCA), which is the prerequisite for all hard magnetic applications. We have investigated one such Heusler alloy Fe$_2$MnSn which shows a high magnetic moment of about 6 $\mu_B$/f.u. Our theoretical calculations showed that both hexagonal D0$_{19}$ and cubic L2$_1$ phases of Fe$_2$MnSn are energetically very close, consistent with the experimental results. Interestingly, the hexagonal phase can be stabilized by manipulating Mn and Sn compositions.

II. METHODS

A. Theory

We performed density functional theory calculations of bulk and thin-film Fe$_2$MnSn Heusler alloy, using the projector augmented-wave method (PAW), implemented in the Vienna ab initio simulation package (VASP) within the generalized-gradient approximation (GGA). The integration method with a 0.05 eV width of smearing is used, along with the plane-wave cut-off energy of 500 eV and energy convergence criteria of 10$^{-2}$ meV for atomic relaxation (resulting in the Hellmann-Feynman forces being less than 0.005 eV/Å), and 10$^{-3}$ meV for the total energy and electronic structure calculations. k-point meshes of 12 × 12 × 12 and 12 × 12 × 16 were used for the Brillouin-zone integration of cubic and hexagonal cells, correspondingly. Convergence of the results with respect to the k-mesh and cut-off energy was tested. For the
calculations of cubic and hexagonal structures, cells with 16 and 8 atoms respectively were used, with periodic boundary condition imposed. For both structures, lattice geometries were fully optimized to obtain equilibrium structures. Some of the results and figures were obtained using the MedeA® software environment. Most of the calculations were performed using Extreme Science and Engineering Discovery Environment (XSEDE) resources located at the Pittsburgh Supercomputing Center (PSC).28

B. Experiment

Polycrystalline ingots with the nominal compositions Fe$_2$MnSn and Fe$_2$Mn$_{1.25}$Sn$_{0.75}$ were prepared by arc-melting pieces of highly pure (at least 99.9%) commercially available metal pellets of Fe, Mn, and Sn under argon atmosphere in water-cooled copper crucible. For better chemical homogeneity, the ingots were re-melted at least five times. In order to compensate the loss of Mn during arc-melting, an additional 2% by weight of Mn was used, which is close to the weight loss measured after melting.29,30 The arc-melted ingots were then vacuum annealed (∼10$^{-7}$ torr) at 800°C for 72 hours followed by quenching in ice-water bath. The crystal structures of the samples were investigated using powder x-ray diffraction (XRD) in a Rigaku MiniFlex600 diffractometer using Cu-Kα x-ray source (λ = 1.54 Å). The temperature and magnetic field dependence of magnetization were measured in a Quantum Design VersaLab magnetometer.

III. RESULTS AND DISCUSSION

A. Theory

Figure 1 (b) shows calculated total and atom-projected density of states (DOS) of bulk Fe$_2$MnSn in cubic L2$_1$ structure as shown in Fig. 1 (a). In the cubic structure with the equilibrium lattice parameter 6.027 Å, the magnetic moment alignment is ferromagnetic. It exhibits high total magnetic moment of 6.04 μ$_B$/f.u. (1.83 μ$_B$/Fe, 2.57 μ$_B$/Mn, and a small induced negative moment of ∼0.14 μ$_B$ on Sn). Further, Fig. 1(b) also indicates that the spin polarization at the Fermi level for the cubic phase of Fe$_2$MnSn is relatively small. The value of P calculated using $P = (N_{\uparrow}(E_F) - N_{\downarrow}(E_F))/(N_{\uparrow}(E_F) + N_{\downarrow}(E_F))$, where $N_{\uparrow}(E_F)$ and $N_{\downarrow}(E_F)$ are the spin-dependent density of states at the Fermi level $E_F$, is ∼27%. The negative sign indicates that the DOS contribution from the minority-spin electrons at the Fermi level is larger than that from the majority-spin electrons. The minority-spin conduction band has an energy gap of ∼0.5 eV.

Figure 2 (a) shows the hexagonal D0$_{19}$ unit cell of Fe$_2$MnSn alloy, and the calculated total and atom-projected density of states for the cell are displayed in Fig. 2(b). In the hexagonal structure with equilibrium lattice parameters $a = 5.389$ Å and $c = 4.310$ Å, the magnetic moment alignment is ferromagnetic. The total magnetic moment of Fe$_2$MnSn in the hexagonal structure is 6.50 μ$_B$/f.u. (2.1 to 2.3 μ$_B$ per Fe and Mn), and a small induced negative moment of...
The temperature and the magnetic field dependence of magnetization of Fe$_2$MnSn and Fe$_2$Mn$_{1.25}$Sn$_{0.75}$ alloys are shown in Figure 4 (a) and (b), respectively. Figure 4 (a) shows the thermomagnetic curves $M(T)$ for the Fe$_2$MnSn and Fe$_2$Mn$_{1.25}$Sn$_{0.75}$ alloys recorded at 1 kOe. The Curie temperatures determined from the lowest points in the corresponding $dM/dT$ versus $T$ curves, as shown in the inset of Figure 4 (a), are 325 K and 475 K for Fe$_2$MnSn and Fe$_2$Mn$_{1.25}$Sn$_{0.75}$ alloys, respectively. There is significant increase in the Curie temperature due to a partial Mn doping for Sn. Figure 4 (b) shows the isothermal magnetization $M(H)$ curves for the Fe$_2$MnSn and Fe$_2$Mn$_{1.25}$Sn$_{0.75}$ alloys recorded at 100 K.

The $M(H)$ curves have very small coercivities and the magnetization of the hexagonal dominant mixed-phase sample saturates at a relatively low field, showing soft magnetic behavior. However, the magnetization of the pure hexagonal sample is not saturated even at 3 T suggesting a high magnetic anisotropy in the sample. We obtained a high magnetic anisotropy of 5.1 Merg/cm$^3$ using the approach-to-saturation method, where the high field data were fitted to $M = M_0(1 - \frac{H}{H_s}) + \chi H$ with $A = \frac{4K_s}{3V_0}$. The parameters $M_0$ and $\chi$ are spontaneous magnetization and high-field susceptibility, respectively. The high-field ($H_s = 3$ T) magnetizations of Fe$_2$MnSn and Fe$_2$Mn$_{1.25}$Sn$_{0.75}$ alloys measured at 100 K are 507 emu/cm$^3$ (3.3 $\mu_B$/f.u.) and 741 emu/cm$^3$ (4.3 $\mu_B$/f.u.), respectively. These experimental values of high-field magnetizations are smaller than the magnetic moments predicted by the first-principle calculations. Although the $M(H)$ curve is saturated for the Fe$_2$MnSn alloy, the structural disorder as observed in the XRD analysis may be causing this decrease in magnetization. In case of the hexagonal

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The calculated value of the spin polarization of Fe$_2$MnSn and Fe$_2$Mn$_{1.25}$Sn$_{0.75}$ alloys is $P = -22\%$, with minority-spin states dominating the Fermi level. Besides, the energy gap of the minority-spin conduction band vanished in the hexagonal structure. We also note that both the cubic and hexagonal structures have very similar equilibrium energies with $-28.97778$ eV/f.u. and $-29.18943$ eV/f.u. being for both the cubic and hexagonal structures respectively. This suggests that both phases may exist in experimental samples.

**B. Experiment**

Figure 3 shows the room temperature x-ray diffraction patterns of Fe$_2$MnSn and Fe$_2$Mn$_{1.25}$Sn$_{0.75}$ Heusler alloys. As shown in Figure 3(a), the XRD pattern contains diffraction peaks from both the cubic and hexagonal (D0$_{0}$ h) phases, hexagonal being the dominant structure. This agrees well with the theoretical prediction. The approximate percentages of cubic and hexagonal phases in Fe$_2$MnSn are 35% and 65%, respectively. The pattern also indicates that the cubic phase is not perfectly ordered. The absence of (111) peak is an indication of the B2 type disorder in the sample. However, with the replacement of some Sn sites with Mn atoms, the hexagonal structure can be stabilized. As shown in Figure 3(b), the XRD pattern of Fe$_2$Mn$_{1.25}$Sn$_{0.75}$ alloy shows a single phase hexagonal D0$_{0}$ phase. In the hexagonal D0$_{0}$ phase of Fe$_2$MnSn, Fe and Mn share the 6h ($\frac{1}{3}, \frac{1}{3}, \frac{2}{3}$), sites with 2/3 and 1/3 occupancy, while Sn occupies the 2d ($\frac{1}{3}, \frac{2}{3}, \frac{1}{3}$) sites.

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phase (Fe$_2$Mn$_{1.25}$Sn$_{0.25}$ alloy), the $M(H)$ curve is not saturated and high-field (higher than 3 T) measurement is necessary to precisely determine the value of saturation magnetization.

IV. CONCLUSIONS

We performed combined theoretical and experimental studies of electronic, magnetic, and structural properties of Fe$_2$MnSn Heusler alloy. Density functional calculations predict that this compound crystallizes in energetically close hexagonal D0$_{19}$ and cubic L2$_1$ phases, which agrees well with the experimental results. Both the Fe$_2$MnSn and Fe$_2$Mn$_{1.25}$Sn$_{0.25}$ alloys are ferromagnetic with high Tc of 325 K and 475 K, respectively. The high-field magnetizations measured at 100 K are 3.3 $\mu_B$/f.u. and 4.3 $\mu_B$/f.u. for Fe$_2$MnSn and Fe$_2$Mn$_{1.25}$Sn$_{0.25}$ alloys, respectively. These values are smaller than the values (~6 $\mu_B$/f.u. for the pure cubic and 6.5 $\mu_B$/f.u for the hexagonal phases) predicted by our calculations. The hexagonal phase (Fe$_2$Mn$_{1.25}$Sn$_{0.25}$ Alloy) shows high value of magnetic anisotropy of 5.1 Merg/cm$^2$ at 100 K. The density functional calculation shows that the cubic phase has an energy gap in the minority-spin conduction band that vanishes in the hexagonal phase. These findings may be interesting for magnetic applications where large saturation magnetization, high Curie temperature, and sizable magnetic anisotropy are desired.

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