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Molecular beam epitaxy growth of Co_2MnSi films on group-IV semiconductors

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1. Introduction

For high-performance spintronic devices, the use of half-metallic materials, where the majority spin channel is metallic while the minority spin channel is semiconductor at the Fermi level, has been regarded as an effective way. Co-based Heusler alloys, Co_2YZ (Y: transition metal, Z: main group element), with an $L2_1$-ordered structure have attracted much attention because of their half-metallic properties and high Curie temperature.[1-3] In particular, very large tunnel magnetoresistance (TMR) ratios have so far been reported for magnetic tunnel junctions (MTJs) with $L2_1$-ordered Co_2MnSi electrodes.[4,5]

Recently, the growth of Co_2MnSi films on Si or Ge by sputtering methods was reported for achieving highly efficient spin injection into group-IV semiconductors.[6] In their works, there was almost no crystallized Co_2MnSi layers in as-grown conditions.[6] Although the amorphous Co_2MnSi layers showed nearly zero magnetic moments, the post-growth annealing at 300~400 ^oC enabled to significantly enhance the magnetic moments.[6] On the other hand, although we have never tried to grow Co_2MnSi films on Si or Ge, we have demonstrated high-quality epitaxial growth of some Heusler alloys, Fe_3Si,[7] Fe_2MnSi,[8] and Co_2FeSi[9,10] by means of low-temperature molecular beam epitaxy (LT-MBE) at less than 200 ^oC.

In this paper, we explore LT-MBE growth of Co_2MnSi films on Si(111) or Ge(111), and investigate the effects of post-growth annealing on magnetic properties.

2. Experimental details

Prior to the growth, we chemically cleaned non-doped Si(111) or Ge(111) substrates using 1 % HF solutions to remove contamination and natural oxide from the surface. The cleaned substrates were loaded immediately into an ultra high vacuum chamber with a base pressure of ~10^{-7} Pa. After a heat treatment conducted at ~450 ^oC for 20 min, the substrate temperature was reduced to ~60 ^oC. After a reflection high-energy electron diffraction (RHEED) patterns of the surface of the Si(111) or Ge(111) substrates indicated an atomically smooth, Co_2MnSi films with a thickness of ~25 nm were directly grown by co-evaporating each element using Knudsen cells.[7-10] During the growth, we performed in-situ RHEED observations for all the samples. Their crystal structures were characterized by transmission electron microscopy (TEM) and nanobeam electron diffraction (NED). Magnetic properties were measured by vibrating sample magnetometer (VSM) at 300 K using a physical property measurement system (Quantum Design).

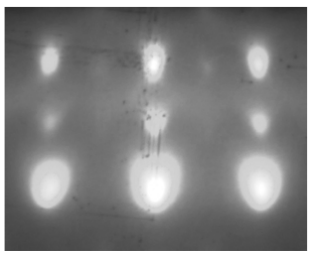

Fig. 1 RHEED patterns of the Co_2MnSi layers, observed along [-211] azimuth.

3. Results and discussion

A representative RHEED pattern of the Co_2MnSi layers grown on Si(111) is displayed in Fig. 1. The RHEED pattern is spotty, indicating the three-dimensional epitaxial growth. Surprisingly, although as-deposited Co_2MnSi films by sputtering methods could not be crystallized,[6] our LT-MBE grown Co_2MnSi films were crystallized even in as-grown conditions. Note that we could not realize two-dimensional epitaxial growth of Co_2MnSi films in any growth conditions. In addition, when we grew Co_2MnSi films on Ge(111), similar features were observed.[10] Interestingly, even for LT-MBE, there exists a large difference in the growth mechanism between Co_2MnSi and other Heusler alloys examined previously in Ref. 7-9.

A cross-sectional TEM image of our LT-MBE grown Co_2MnSi film is shown in Fig. 2. The insets show NED patterns of the Co_2MnSi layers measured at point #1 and #2, as denoted in the figure. Although we can confirm no reaction layers near the heterointerface between Co_2MnSi and Si, our Co_2MnSi films consist of single- (point #1) and poly- (point #2) crystalline phases. In the NED pattern at point #1, we can see superlattice reflections, caused by the presence of an $L2_1$-ordered structure (solid circles), togeth-

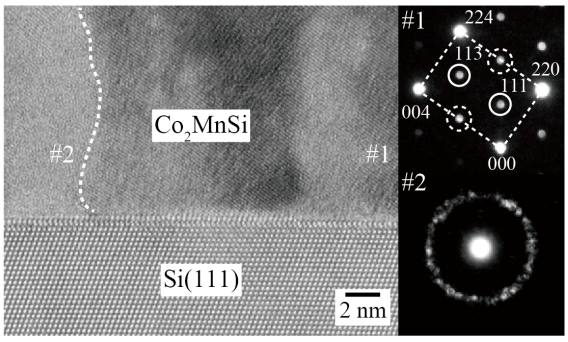

Fig. 2 Cross-sectional TEM images near the Co_2MnSi/Si(111) interface. The insets show NED patterns of the Co_2MnSi layers. The zone axis of the incident electrons is parallel to [1-10] directions.

er with superlattice reflections of the ordered $B2$+$L2_1$ structures (dotted circles). Namely, epitaxial Co_2MnSi layer with an $L2_1$-ordered structure was obtained even in an as-grown condition. Almost the same features were seen for the growth on Ge(111). These mean that the use of the LT-MBE techniques is a key to form the crystalline Co_2MnSi films on Si or Ge in as-grown conditions. Further exploration is required to obtain highly ordered Co_2MnSi films.

Figures 3(a) and 3(b) show field-dependent magnetization (M-H) curves of our as-grown Co_2MnSi layers on Si(111) and Ge(111). The magnetic moment (M_S) of the Co_2MnSi layers on Si(111) or Ge(111) is ~2.8 μ_B/f.u. or ~1.6 μ_B/f.u., respectively, nearly or less than half that of bulk Co_2MnSi (~5 μ_B/f.u.)[3]. Since there are some single-crystalline phases in the grown Co_2MnSi films in as-grown conditions, M_S values for our Co_2MnSi films were higher than those for as-deposited Co_2MnSi films by sputtering methods.[6] Since our as-grown Co_2MnSi films also contain poly-crystalline phases, the coercivity is very large (> 150 Oe), which deviates from the characteristics of highly ordered Heusler alloys.[2,3,7-10]

Next, we conducted post-growth annealing at various annealing temperatures (T_A) in vacuum. We summarize M_S as a function of T_A in Fig. 3(c). With increasing T_A, we observed an enhancement in M_S for both samples. Here we also plotted representative M-H curves of the annealed Co_2MnSi layers in Figs. 3(a) and 3(b). The coercivity is significantly reduced. These results indicate that the post-growth annealing promoted the crystallization and $L2_1$-ordering, leading to the enhancement in M_S and the reduction in the coercivity. Above a certain T_A, M_S monotonically decreases with increasing T_A, probably due to the formation of nonmagnetic phases by interfacial reaction. Even for the use of MBE techniques, post-growth annealing is required to obtain highly ordered Co_2MnSi films.

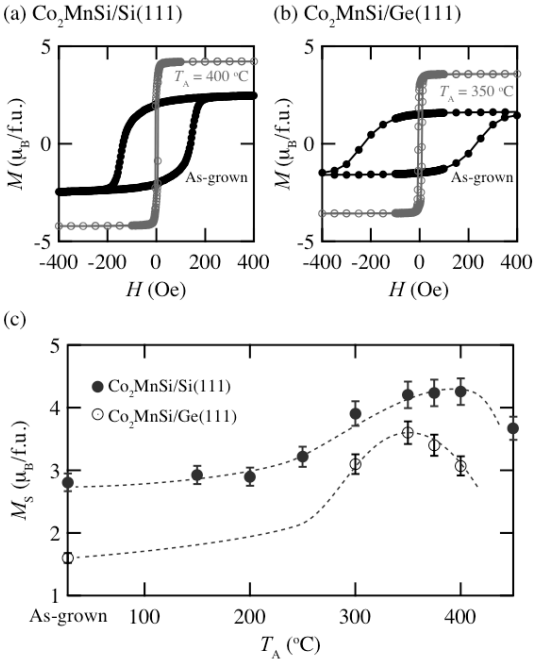

Fig. 3 M-H curves of the as-grown and annealed Co_2MnSi layers grown on (a) Si(111) and (b) Ge(111) at 300 K. (c) M_S as a function of T_A.

4. Conclusions

We explored the crystal growth of Co_2MnSi on Si(111) or Ge(111) by means of low-temperature molecular beam epitaxy. The as-grown Co_2MnSi films consist of mixed phases with $L2_1$-ordered and poly-crystalline structures. Post-growth annealing works effectively to promote the crystallization of the Co_2MnSi films, leading to an enhancement in magnetic moment. These results are quite different from those for other Heusler alloys examined in our previous works. It is very important to explore and optimize the growth conditions according to a kind of alloys.

Acknowledgements

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