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# 1 Experimental inspection of a computationally-designed NiCrMnSi 2 Heusler alloy with high Curie temperature

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13  
14 Nowadays advanced magnetic tunnel junction applications demand very high tunnel magnetoresistance at  
15 room temperature, thus it is quite important to explore high Curie temperature  $T_c$  half-metallic Heusler  
16 alloys. In this article first-principles calculation unveiled that NiCrMnSi has  $T_c$  of 1200 K comparable to  
17 that of the traditional Co<sub>2</sub>MnSi Heusler alloys, even though it does not contain Co element. In addition,  
18 we examined whether NiCrMnSi Heusler phase films can be obtained by a magnetron sputtering on MgO  
19 substrates. The results of the structural analysis and first-principles calculations indicated that NiCrMnSi  
20 Heusler phase is metastable. A possible route to obtain metastable NiCrMnSi Heusler alloy is to utilize  
21 appropriate templates.

## 22 1. Introduction

23 Magnetic tunnel junctions (MTJs) are currently being widely used for various com-  
24 mercial applications, such as storage, sensor, and memory. More recently, advanced  
25 computing technologies are emergent and demand MTJs exhibiting much higher tun-  
26 nel magnetoresistance (TMR) ratio at room temperature.<sup>1-3)</sup> Large TMR ratios can  
27 be obtained in MTJs with a use of electrodes of half-metallic ferromagnets which are  
28 magnetic metals having a band gap at the Fermi level for their minority spin state.<sup>4)</sup> A  
29 Co<sub>2</sub>-based Heusler alloy is one of the candidates of half-metals and attracted much at-  
30 tention to date.<sup>5)</sup> Among several kinds of Co<sub>2</sub>-based Heusler alloys, a promising one for

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1 MTJs is  $\text{Co}_2\text{MnSi}$ .<sup>6)</sup> Indeed, MTJs with electrodes of  $\text{Co}_2\text{MnSi}$  or of the ones with Mn  
2 partially substituted by Fe showed the record of the TMR ratio of about 2600% at low  
3 temperature.<sup>7)</sup> However, this TMR ratio is significantly reduced at room temperature.<sup>7)</sup>  
4 The reduction of the TMR ratio with elevating temperature is due to thermal fluctua-  
5 tion of a magnetic moment,<sup>8-10)</sup> hence it can be suppressed with use of half-metals with  
6 higher Curie temperatures  $T_c$ .

7 A numerous elemental combinations in Heusler alloys enabled us to find new com-  
8 positions showing unconventional electronic structures, such as a spin gapless semi-  
9 conductor.<sup>11)</sup> Furthermore, computation-based exploration are much progressed.<sup>12-15)</sup>  
10 However, there are yet no computational reports focusing on half-metallic Heusler al-  
11 loys with high  $T_c$  beyond those of  $\text{Co}_2\text{MnSi}$  and the derivatives.

12 As a first step of computational/experimental explorations for high  $T_c$  half-metallic  
13 Heusler alloys, we theoretically and experimentally studied NiCrMnSi equiatomic qua-  
14 ternary alloys in this article. The *ab-initio* calculations unveiled, for the first time, that  
15 the NiCrMnSi Heusler alloy has attractive properties; such as a small net magnetic  
16 moment due to the ferrimagnetic spin structure, high  $T_c$ , and large spin polarization,  
17 even though elemental compositions are different from  $\text{Co}_2$ -based Heusler alloys. We  
18 also examined whether film growth of NiCrMnSi is possible using a magnetron sput-  
19 tering technique. The experiments indicated that NiCrMnSi films are decomposed into  
20 other alloys/compounds without crystallizing into Heusler phase. This phase separation  
21 observed in the experiments is explained with the formation energies for NiCrMnSi and  
22 the others, and indicating that NiCrMnSi Heusler phase is rather metastable. A possible  
23 route to obtain the Heusler phase film of NiCrMnSi is discussed.

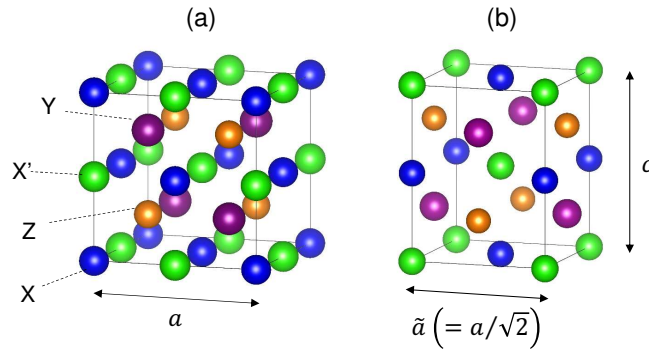
## 24 **2. First-principles calculations**

25 *Ab-initio* calculation of NiCrMnSi has been performed using Vienna Ab Initio Simu-  
26 lation Package,<sup>16)</sup> in combination with the projector augmented wave method.<sup>17)</sup> For  
27 the exchange correlation potential, we used the generalized gradient approximation  
28 (GGA).<sup>18)</sup> The energy cutoff of 500 eV has been set for the plane waves, and the fi-  
29 nal energies have been calculated with a  $k$ -mesh of  $8 \times 8 \times 8$  for the cubic case and a  
30 comparable number of  $k$ -points used for the tetragonal case. The energy and the force  
31 tolerance were 10  $\mu\text{eV}$  and 10  $\text{meV}/\text{\AA}$ , respectively.

32 Magnetic interactions and the detail insight of magnetism have been probed us-  
33 ing Green's function based spin-polarized relativistic Korringa-Kohn-Rostoker method

**Table I.** Calculated formation energies  $\Delta E_{XX'YZ}$  and lattice constant  $a$  for cubic and tetragonal Ni-Cr-Mn-Si Heusler alloys with different types.  $c/a$  is the axial ratio of the tetragonal unit cell.

XX'YZ	structure	$\Delta E_{XX'YZ}$ (eV/f.u.)	$a$ (Å)	$c/a$
NiCrMnSi	cubic	-1.033	5.710	
	tetragonal	-1.151	5.285	1.270
MnNiCrSi	cubic	-0.974	5.681	
	tetragonal	-1.118	5.213	1.254
MnCrNiSi	cubic	-0.402	5.658	
	tetragonal	-0.794	5.046	1.457



**Fig. 1.** Illustration of a crystal structure of  $Y$  ordered equiatomic Heusler alloy  $XX'YZ$  with (a) cubic ( $a = c$ ) or (b) tetragonal ( $a \neq c$ ) lattice. Cubic  $XX'YZ$  Heusler structure has a space group of  $F\bar{4}3m$  (no. 216) and tetragonal lattice has a space group of  $I\bar{4}m2$  (no. 119).

1 (SPR-KKR) as implemented in the programme package.<sup>19)</sup> We calculate the Heisenberg  
 2 exchange coupling constant within a real space approach as proposed by Liechtenstein *et*  
 3 *al.*<sup>20)</sup> Furthermore,  $T_c$  has been calculated in terms of the Heisenberg exchange coupling  
 4 constant, within mean field approximation. For the self-consistent-field calculations we  
 5 use the full-potential method and GGA for the exchange correlation potential. We use  
 6 824 irreducible  $k$ -points for the Brillouin zone integration and an angular momentum  
 7 expansion up to 3 has been used for each atom. 90 energy points on the complex energy  
 8 path were used.

9 We evaluated the formation energy for the three types of ordered structure of  
 10 NiCrMnSi with cubic or tetragonal structures, as shown in Fig. 1. Here the formation  
 11 energy  $\Delta E_{XX'YZ}$  is defined as,

$$\Delta E_{XX'YZ} = E_{XX'YZ} - [E_X + E_{X'} + E_Y + E_Z], \quad (1)$$

12 where  $E_{XX'YZ}$  denotes the total energy of the Heusler alloy  $XX'YZ$ , and  $E_X$ ,  $E_{X'}$ ,  $E_Y$

**Table II.** Physical properties of tetragonal NiCrMnSi predicted from the first principles; the Curie temperature  $T_c$ , the spin polarization  $P$ , the basal plane lattice constant  $a$ , the axial ratio  $c/a$ , and the total magnetic moment  $m_{\text{tot}}$ . The values for Co<sub>2</sub>MnSi similarly calculated from the first principles are also shown for comparison. Values in the parentheses correspond to the partial magnetic moments of Ni, Cr, and Mn atom for NiCrMnSi and those of Co and Mn atom for Co<sub>2</sub>MnSi, respectively.

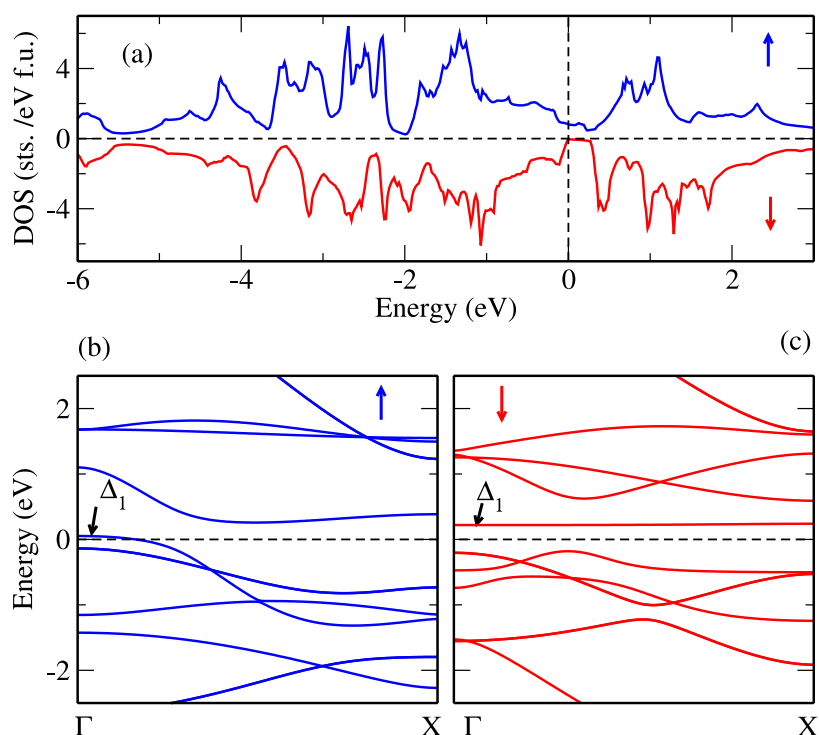
Material	$T_c$ (K)	$P$ (%)	$a$ (nm)	$c/a$	$m_{\text{tot}}$ ( $\mu_B$ /f.u.)
NiCrMnSi	1200	88	0.528	1.27	0.95 (0.13, -1.79, 2.61)
Co <sub>2</sub> MnSi	1204	100	0.563	1.00	5.00 (1.02, 2.99)

1 and  $E_Z$  are those of elemental solids, X, X', Y and Z, respectively. Then we found  
2 the tetragonal type of NiCrMnSi is the most stable in terms of the formation energy,  
3 as shown in Table I. The predicted physical properties are summarized in Table II.  
4 We also calculated the values for Co<sub>2</sub>MnSi from the first principles and those are also  
5 shown in the Table II for comparison. The Curie temperature of NiCrMnSi is as high  
6 as that of Co<sub>2</sub>MnSi, which stems from the large antiferromagnetic exchange interaction  
7 between magnetic moments for Cr and Mn. Note that the mean-field approximation  
8 usually tends to overestimate the Curie temperature since it neglects the thermal fluc-  
9 tuation of magnetic moments. Indeed, the Curie temperature experimentally observed  
10 for Co<sub>2</sub>MnSi was 990 K.<sup>21)</sup> Total magnetic moment is close to 1  $\mu_B$  and the spin polar-  
11 ization is close to unity, because the Fermi level is located near the top of the valence  
12 band edge for the minority spin state, as shown in Fig. 2(a). Importantly, the  $\Delta_1$  band  
13 dispersion along the [001] axis is located across the Fermi level for the majority spin  
14 state and no  $\Delta_1$  bands are located at the Fermi level for the minority spin state, as  
15 shown in Figs. 2(b) and 2(c), respectively. This means that the  $\Delta_1$  band which de-  
16 termines the TMR ratio in MgO-based MTJs is fully spin polarized, hence the large  
17 TMR ratio is expected, as theoretically predicted and experimentally observed in bcc  
18 Fe(-Co)/MgO MTJs.<sup>22-26)</sup>

### 19 3. Experiments

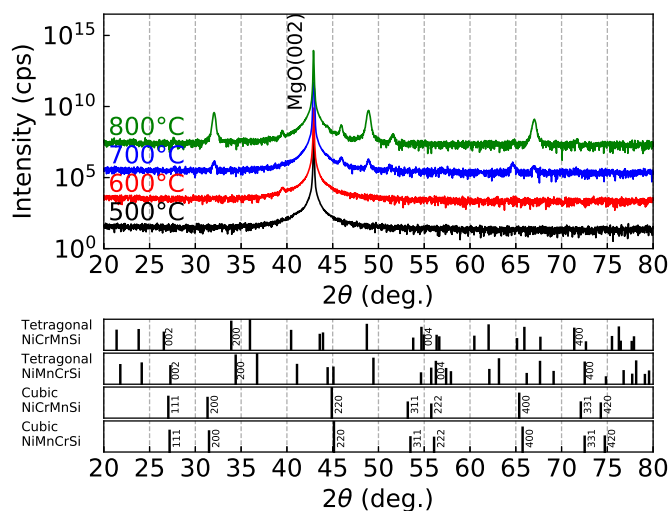
#### 20 3.1 Experimental procedures

21 All samples were prepared by a magnetron sputtering system with a base pressure of  
22  $2 \times 10^{-7}$  Pa, as previously used in different Heusler alloy films.<sup>27-30)</sup> The single crystal  
23 MgO(001) substrates were cleaned by ultrasonic cleaning in an acetone and an ethanol  
24 prior to being introducing into the load-lock chamber. Sample stacking structure was



**Fig. 2.** Electronic structure calculated from the first principles for a tetragonal NiCrMnSi. (a) The density-of-states, and the band dispersion along the [001] axis for (b) majority and (c) minority spin states.

1 MgO(001) substrate /NiCrMnSi(100 nm)/Ta(3 nm). The substrates were thermally  
 2 flushed at 700°C in the chamber to remove residual organic substances and water.  
 3 NiCrMnSi layers were deposited by a co-sputtering technique using NiCr and MnSi alloy  
 4 targets. Powers were 100 W, 30 W, 26 W and 60 W in the Cr, NiCr, MnSi and Ta targets,  
 5 respectively. The composition of Ni : Cr : Mn : Si in the films was evaluated as 25.9 :  
 6 29.5 : 23.0 : 21.6 (at%) by an inductively coupled plasma mass spectrometry (ICP-  
 7 MS). The 100-nm-thick NiCrMnSi layers were deposited at a substrate temperature  
 8  $T_s$  of 500–800°C. Finally, Ta capping layers were deposited at room temperature. The  
 9 crystalline structure of the samples was measured by an out-of-plane X-ray diffraction  
 10 (XRD) with Cu  $K_\alpha$  radiation (wavelength of 0.15418 nm) and a transmission electron  
 11 microscope (TEM). Magnetic property was measured using a polar magneto-optical  
 12 Kerr effect (p-MOKE) with a maximum magnetic field of 2 T and a laser wavelength  
 13 of approximately 400 nm. Magnetization measurements were also carried out using a  
 14 vibrating sample magnetometer (VSM) with a maximum in-plane magnetic field of 2



**Fig. 3.** Out of plane XRD patterns of NiCrMnSi films with different deposition temperatures. Theoretical diffraction patterns for the NiCrMnSi and NiMnCrSi Heusler phases with cubic and tetragonal structures are shown at the bottom.

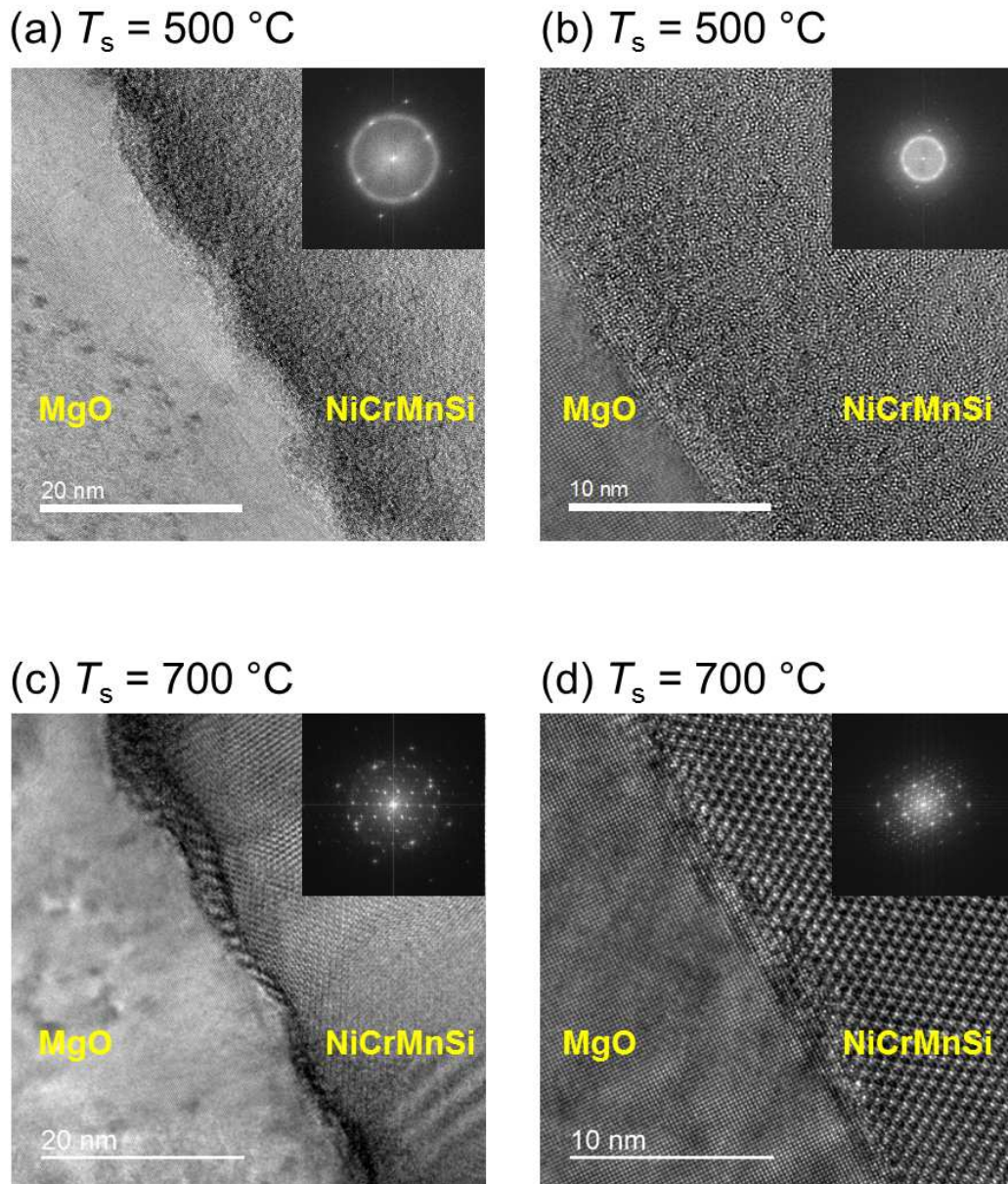
1 T.

## 2 3.2 Experimental results

3 The out-of-plane XRD patterns for the NiCrMnSi films on MgO substrates with dif-  
 4 ferent deposition temperatures are shown in Fig. 3. The powder diffraction patterns  
 5 for the NiCrMnSi Heusler phase with cubic and tetragonal structures are theoretically  
 6 simulated and are also plotted at the bottom for comparison. The diffraction peaks  
 7 were observed for the samples with  $T_s \geq 700^\circ\text{C}$  and the peak intensity increased with  
 8 increasing the annealing temperature. However, the peak positions were not consistent  
 9 with those from the cubic or tetragonal phase. Therefore, the other phases might be  
 10 formed in these samples. However, there are many candidates consisting each element  
 11 corresponding to these peaks and it is difficult to identify the formed phases from these  
 12 XRD patterns.

13 To consider the phase formed in these samples, TEM observation was performed.  
 14 Figures 4(a) and 4(b) show the TEM images for NiCrMnSi films deposited on MgO  
 15 substrates at  $500^\circ\text{C}$  with low and high magnification, respectively. Figures 4(c) and  
 16 4(d) similarly show those for the films deposited at  $700^\circ\text{C}$  with low and high magni-  
 17 fication, respectively. The corresponding diffraction patterns are included as insets in  
 18 these figures. In the case for the sample deposited at  $500^\circ\text{C}$ , some short-range crystal-  
 19 lization phases were observed and the ring pattern for the diffraction images indicated

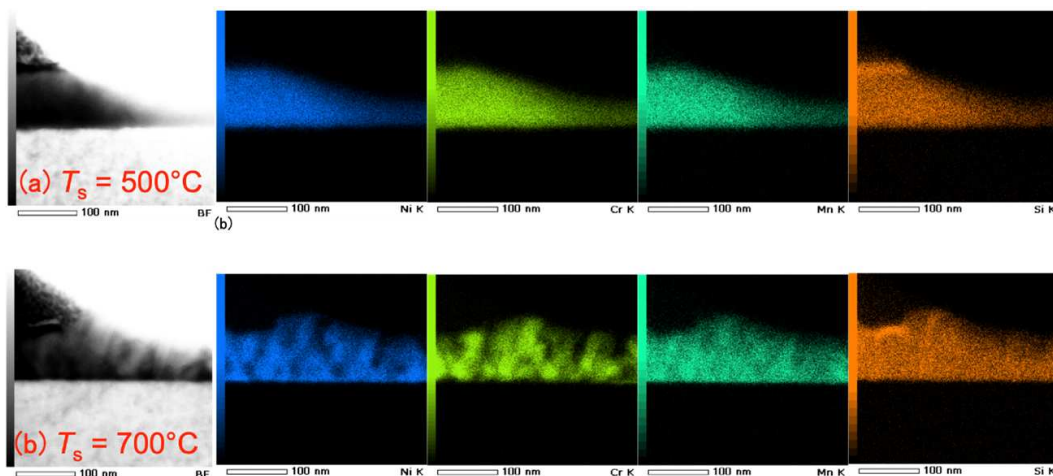




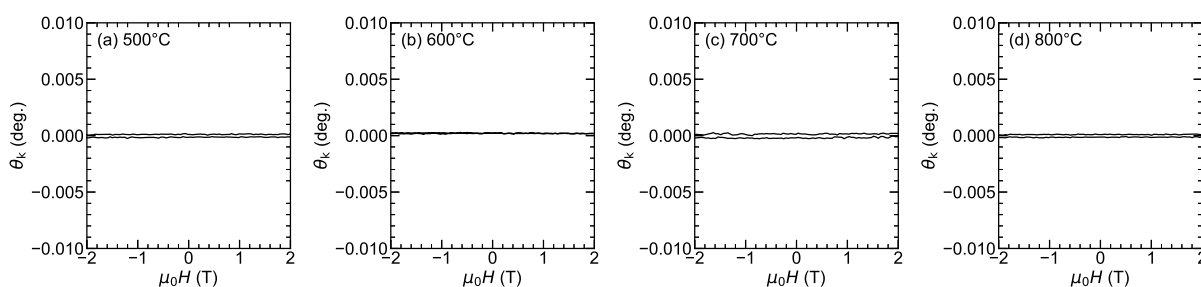
**Fig. 4.** TEM images of NiCrMnSi deposited at 500°C and 700°C. (a) Low and (b) high magnification images for NiCrMnSi deposited at 500°C. (c) Low and (d) high magnification images for NiCrMnSi deposited at 700°C. Insets are the corresponding selected beam electron diffraction pattern.

1 the formation of polycrystalline. Because of the short-range crystallization, XRD mea-  
2 surements showed no diffraction peak in Fig. 3. On the other hand, in the case for the  
3 sample deposited at 700°C, the grain boundaries are observed clearly for the images  
4 with low magnification. It suggested the grain sizes are larger than those for the sample  
5 deposited at 500°C. Furthermore, the diffraction spots were observed and suggested





**Fig. 5.** Element mapping images by TEM-EDX for NiCrMnSi samples deposited at (a) 500°C and (b) 700°C. Left images are TEM images. Blue, yellow green, and orange data points corresponding to the Ni, Cr, Mn and Si elements, respectively.



**Fig. 6.** p-MOKE results for the NiCrMnSi deposited at (a) 500°C, (b) 600°C, (c) 700°C, and (d) 800°C.

1 that multiple crystalline phases were formed as seen as their overlapped patterns. Ob-  
 2 servation agrees with the fact that many diffraction peaks were observed in the XRD  
 3 measurements for the sample deposited at 700°C in Fig. 3.

4 For further analysis, the elemental mapping was performed by energy dispersive X-  
 5 ray spectrometry (EDX) in the TEM apparatus. Figures 5(a) and 5(b) show the cross-  
 6 sectional TEM images and elemental mapping images for NiCrMnSi films deposited  
 7 at 500°C and 700°C, respectively. Left figures are cross-sectional TEM images and the  
 8 other four figures are elemental mapping images. Blue, yellow-green, green, and orange  
 9 correspond to the detected Ni, Cr, Mn, and Si elements, respectively. For the sample  
 10 deposited at 500°C, all four elements were homogeneously distributed in a whole region.  
 11 However, non-uniform distributions for Ni and Cr atoms were observed and these are  
 12 complementary for the sample deposited at 700°C. It suggests that the Ni-rich phase

1 and the Cr-rich phase were spatially separated in this sample.

2 The magnetic properties of NiCrMnSi films were measured by using the p-MOKE.  
 3 Figure 6 shows the out-of-plane MOKE curves for the NiCrMnSi films with different  
 4 deposition temperatures. No hysteresis loop was observed for all the samples even at  
 5 the deposition temperature of 800°C. Some samples were also measured using VSM,  
 6 however, no hysteresis loop was observed. These results suggested that all samples in  
 7 this study showed paramagnetism or antiferromagnetism at room temperature.

#### 8 4. Discussion

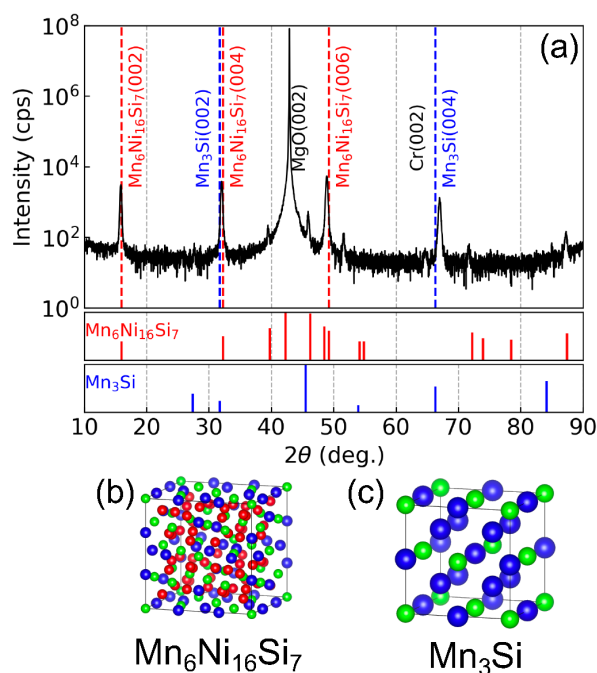
9 To investigate which phases were formed, the out-of-plane XRD pattern with the wide  
 10 range from 10° to 90° for the sample deposited at 800°C is shown in Fig. 7(a). From the  
 11 XRD analysis and TEM observation, (001)-orientated Mn<sub>6</sub>Ni<sub>16</sub>Si<sub>7</sub> and Mn<sub>3</sub>Si may be  
 12 formed as dominants phases in the samples. Figures 7(b) and 7(c) show the crystalline  
 13 lattices of Mn<sub>6</sub>Ni<sub>16</sub>Si<sub>7</sub> and Mn<sub>3</sub>Si, respectively. Mn<sub>6</sub>Ni<sub>16</sub>Si<sub>7</sub> and Mn<sub>3</sub>Si are antiferro-  
 14 magnets with the Neel temperatures of  $\simeq 200$  K<sup>31)</sup> and 23 K,<sup>32)</sup> respectively. Therefore,  
 15 this analysis is consistent with the magnetic measurements which showed no hysteresis  
 16 loop in Fig. 6. In addition to the above mentioned phases, there exist minor phases.  
 17 For example, some peaks as appeared in Fig. 3 and Fig. 7(a) cannot be attributed to  
 18 Mn<sub>6</sub>Ni<sub>16</sub>Si<sub>7</sub>, and Mn<sub>3</sub>Si. Those unknown phases may be Cr-rich alloys or compounds  
 19 and minor residues.

20 The phase stability can be discussed by the difference  $\delta E$  in the formation energy  
 21 of NiCrMnSi and alloys/compounds decomposed. When we assume that the above-  
 22 mentioned unknown phases are Cr<sub>3</sub>Si and Si,  $\delta E$  is evaluated as,

$$\delta E = \Delta E_{\text{NiCrMnSi}} - \left[ \frac{1}{16} \Delta E_{\text{Mn}_6\text{Ni}_{16}\text{Si}_7} + \frac{5}{24} \Delta E_{\text{Mn}_3\text{Si}} + \frac{1}{3} \Delta E_{\text{Cr}_3\text{Si}} + \frac{1}{48} \Delta E_{\text{Si}} \right].$$

23 Here,  $\Delta E_{\text{Mn}_6\text{Ni}_{16}\text{Si}_7}$ ,  $\Delta E_{\text{Mn}_3\text{Si}}$ ,  $\Delta E_{\text{Cr}_3\text{Si}}$ , and  $\Delta E_{\text{Si}}$  are the formation energies of bulk  
 24 Mn<sub>6</sub>Ni<sub>16</sub>Si<sub>7</sub>, Mn<sub>3</sub>Si, Cr<sub>3</sub>Si, and Si, respectively which are found to be  $-12.686$  eV/f.u.,  
 25  $-1.334$  eV/f.u.,  $-1.436$  eV/f.u., and  $0$  eV/f.u. After substituting these values in the  
 26 above equation, we obtain  $\delta E = 0.396$  eV/f.u. The positive value of  $\delta E$  means that the  
 27 Heusler phase of NiCrMnSi is not stable at ground state and tends to be decomposed  
 28 into the others in the bulk form. Thus this analysis reasonably accounts the experimental  
 29 observations.

30 Above discussion implies that the Heusler phase of NiCrMnSi is rather metastable.



**Fig. 7.** (a) Wide range out of plane XRD pattern of NiCrMnSi deposited at 800°C. Theoretical diffraction patterns for  $\text{Mn}_6\text{Ni}_{16}\text{Si}_7$  and  $\text{Mn}_3\text{Si}$  are shown at the bottom. Cartoons of the crystal lattice of (b)  $\text{Mn}_6\text{Ni}_{16}\text{Si}_7$  and (c)  $\text{Mn}_3\text{Si}$ .

1 In general, metastable phases of alloy films can be obtained when those are grown on  
 2 an appropriate template with optimized growth conditions such as film thickness and  
 3 growth temperatures. In particular, relatively low temperature growth on a specific alloy  
 4 template would be effective, as well known in tetragonal Mn-based Heusler alloys and  
 5 those derivatives, for example.<sup>33–40)</sup> Thus, it would be crucial to find good templates  
 6 for obtaining the Heusler phase of NiCrMnSi, which is left as a future subject.

## 7 5. Summary

8 We theoretically and experimentally studied on NiCrMnSi equiatomic Heusler alloys.  
 9 The first principles calculation predicted that the tetragonal NiCrMnSi was energeti-  
 10 cally favorable compared with the cubic phase and possessed the high Curie temperature  
 11 and the fully spin-polarized electronic band along [001] directions. NiCrMnSi equiatomic  
 12 alloy films were fabricated by magnetron sputtering. The XRD measurements indicated  
 13 that several phases were formed for the samples deposited at high deposition tempera-  
 14 tures. The TEM observation showed that the Ni-rich and Cr-rich phases were spatially  
 15 separated in those films. The p-MOKE and VSM measurements showed no magnetic  
 16 hysteresis loop, which suggested that all samples have paramagnetism or antiferromag-

1 netism at room temperature. This phase separation was also theoretically explained  
2 in terms of the formation energies of NiCrMnSi and the other decomposed alloys and  
3 compounds. A possible route to obtain the NiCrMnSi single phase Heusler alloys as the  
4 metastable phase is to utilize more appropriate template to promote the crystallization  
5 of NiCrMnSi Heusler alloys at relatively lower temperature.

#### 6 **acknowledgement**

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