Neutron Diffraction Study of Heusler Type Alloy $\text{Mn}_{0.47}\text{V}_{0.28}\text{Al}_{0.25}$

By Hiroshi Itoh*, Takurô Nakamichi**,
Yasuo Yamaguchi** and Noriaki Kazama**

Crystal and magnetic structures of an annealed Mn-V-Al ternary alloy were investigated by powder neutron diffraction. Neutron diffraction measurements confirmed clearly that the Mn-V-Al alloy specimen with approximate composition Mn_2VAl annealed at 1023 K for 48 h has the Heusler (L_{21})-type structure. Our studies on the annealed alloy specimen, $\text{Mn}_{1.88}\text{V}_{1.12}\text{Al}_{1.00}$, showed that Mn atom occupies preferentially the simple cubic Mn sublattice $\alpha(\text{A}+\text{C})$, while V and Al atoms occupy the B and D sublattices, respectively, in the *bcc* structure composed of four equivalent *fcc* sublattices A, B, C and D. It was suggested strongly that Mn atom has the magnetic moment $(1.5 \pm 0.3)\mu_B$ in the sublattice α and couples antiferromagnetically with V atom which has the magnetic moment $0.9\mu_B$ in the B sublattice.

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I. Introduction

The phase constitution in the Mn-V-Al ternary system was investigated extensively by Kopp and Wachtel⁽¹⁾, and a very wide *bcc* disordered structure was found. Our precise X-ray measurements showed that there are two kinds of ordered structures in this *bcc* Mn-V-Al ternary alloy system⁽²⁾⁽³⁾.

The magnetic properties of these alloys vary sensitively with composition and heat treatment, corresponding to phase changes due to atomic ordering. Furthermore, several alloys showed thermomagnetic curves typical of ferrimagnetism. They have a large saturation magnetization near the Curie temperature rather than at lower temperatures⁽⁴⁾. This fact shows that there is a certain kind of antiferromagnetic alignment of magnetic moments in

this alloy system.

The most important result in our previous works⁽³⁾⁽⁴⁾ is that the stoichiometric composition Mn_2VAl has the Heusler(L_{21})-type structure. This result was suggested by anomalous dispersion of X-rays⁽⁵⁾, which enhances the difference in the effective atomic scattering amplitudes between Mn and V. The L_{21} structure is one of the ordered structures based on the *bcc* structure, which can be divided into four equivalent *fcc* sublattices A, B, C and D illustrated in Fig. 1. In the usual Heusler alloy X_2YZ such as Cu_2MnAl , X atom occupies the A and C sublattices forming a simple cubic sublattice α , and Y and Z atoms occupy the B and D sublattices, respectively. On the other hand, in the alloy Mn_2VAl , it was concluded from the X-ray measurements that Mn atom occupies the A and C sublattices⁽³⁾. The alloy is of a new Heusler type, in which Mn atom loses its preference location in the B sublattice known generally for nearly thirty other Heusler type alloys X_2MnZ .

It is very valuable to confirm the above conclusion by neutron diffraction measurements. This is because it gives a remarkable difference between the nuclear cross sections of Mn and V and also gives the distinct arrangement of the atoms more precisely.

In the present paper, we report results of

* Research institute for Iron, Steel and Other Metals, Tohoku University, Sendai. Present Address: Electronic Engineering Research Laboratory, Pioneer Electronic Corporation, Tokorozawa-shi, Saitama 359, Japan.

** Research Institute for Iron, Steel and Other Metals, Tohoku University, Sendai 980, Japan.

The units used in this paper are converted to SI unit as follows: $1 \text{ \AA} = 10^{-10} \text{ m}$, $1 \text{ Oe} = 79.6 \text{ A/m}$ and $1 \mu_B = 1.653 \times 10^{-29} \text{ Wb} \cdot \text{m}$.

Fig. 1 The doubly ordered *bcc* structure consisting of four equivalent *fcc* sublattices A, B, C and D. In the Heusler ($L2_1$)-type structure and CsCl($B2$)-type structure, the simplification of the following representations may be used:

$L2_1$; $A+C \rightarrow \alpha$, $B \rightarrow \beta$, $D \rightarrow \gamma$; $B2$; $A+C \rightarrow I(\alpha)$, $B+D \rightarrow II(\beta+\gamma)$.

powder neutron diffraction measurements for the alloy specimen $Mn_{0.47}V_{0.28}Al_{0.25}$ having nearly stoichiometric composition Mn_2VAl . The crystal and magnetic structures of this alloy specimen are determined, and the origin of the antiferromagnetic interaction between the magnetic elements is discussed.

II. Specimen Preparation and Experimental Procedure

The specimen was prepared by arc-melting of electrolytic manganese, vanadium chip and super-purified aluminum rod using the same procedure as in previous papers^{(2)–(4)}. After the ingot was homogenized at 1473 K for 48 h and then dropped quickly into ice water. The quartz tube was broken immediately to expose the ingot to the ice water (quenched condition). This ingot was sealed again in another quartz tube, annealed at 1023 K for 48 h, cooled slowly down to 723 K at the rate of 275 K/h and finally cooled slowly down to room temperature (annealed condition). Chemical analysis showed the composition to be $Mn_{0.47}V_{0.28}Al_{0.25}$.

This doubly annealed ingot was then crushed and milled into fine powder. This powder was pressed in a cylindrical form 0.03 m in diameter

and 0.028 m in length. This pressed specimen was sealed in a quartz tube and again annealed at the same annealed condition as described above.

The TOG neutron diffractometer installed at the JRR-3 Reactor in JAERI was used for the neutron diffraction measurements. The cylindrical powder specimen, which was enclosed in a thin aluminum foil, was placed between the pole pieces of an electromagnet so that its cylindrical axis was parallel to the direction of magnetic field. The electromagnet was mounted on a goniometer, to permit the θ - 2θ scanning. Neutron diffraction measurements were performed using a neutron beam of 0.102 nm wavelength with and without external magnetic field at room temperature. Its magnetic field was 672 kA/m, which was enough to saturate the magnetization of the specimen⁽⁴⁾.

III. Experimental Results

Neutron diffraction patterns are shown in Figs. 2(a) and 2(b). The lower pattern (a) corresponds to the nuclear scattering contribution for the specimen, when the external magnetic field was applied to the direction of the scattering vector. The upper pattern (b) representing the magnetic scattering contribution is the difference between the neutron diffraction patterns of the specimen with and without magnetic field, i.e. $I_{H=0} - I_{H=672 \text{ kA/m}}$.

In the nuclear scattering contribution (a), all of the scattering peaks could be indexed in a doubly ordered superlattice structure consisting with the previous X-ray measurements^{(2)–(4)}. The *bcc* fundamental peaks restricted to $h+k+l=4n$, such as 220 and 400 are rather weak, and strong peaks are found at the 200 and 222 positions, which belong to the $h+k+l=4n+2$ group. Moreover, the 111 and 311 peaks are observed distinctly. These peaks belong to $h, k, l = \text{all odd}$ group and relates essentially to a $L2_1$ like structure. These results are discussed in the following section.

In the magnetic scattering contribution, the 200 peak is strongest and other peaks are all very weak compared with the 200 peak as seen in Fig. 2(b), and one must recognize the existence of the weak but important magnetic

Fig. 2 Neutron diffraction patterns of the alloy specimen $\text{Mn}_{0.47}\text{V}_{0.28}\text{Al}_{0.25}$. The lower pattern (a) represents the nuclear scattering contribution and the upper pattern (b) the magnetic scattering contribution.

111 peak, which offers some interesting but troublesome information concerning the magnetic structure of this alloy. This will also be discussed in the following section.

IV. Reliability Factor and Structure Determination

The neutron diffraction intensities are given by the expressions of I_{hkl}^N for the nuclear scattering contribution and I_{hkl}^M for the magnetic scattering contribution, neglecting the absorption factor⁽⁶⁾,

$$I_{hkl}^N/K = j \cdot \sin^{-2} 2\theta \cdot e^{-2W} |F_{hkl}^N|^2 \quad (1)$$

and

$$I_{hkl}^M/K = (2/3)j \cdot \sin^{-2} 2\theta \cdot e^{-2W} |F_{hkl}^M|^2, \quad (2)$$

where K is a constant relating to the apparatus, j the multiplicity factor, θ the Bragg angle, e^{-2W} the Debye-Waller factor and F_{hkl}^N and F_{hkl}^M the nuclear and magnetic structure factors, respectively. The suffixes N and M mean the

nuclear and magnetic scattering contributions, respectively.

To interpret the nuclear scattering patterns shown in Fig. 2(a), we assume the crystal structure having the equivalent *fcc* sublattices A, B, C and D, as illustrated in Fig. 1. In such a structure, the structure factor designated in eq. (1) can be written as follows:

$h, k, l = \text{all odd}:$

$$F_{hkl}^N = 4\{(f_A - f_C)^2 + (f_B - f_D)^2\}^{1/2}, \quad (3)$$

$h, k, l = \text{all even}:$

$$h + k + l = 4n + 2: F_{hkl}^N = 4|(f_A + f_C) - (f_B + f_D)|, \quad (4)$$

$$h + k + l = 4n: F_{hkl}^N = 4|(f_A + f_C) + (f_B + f_D)|, \quad (5)$$

$h, k, l = \text{otherwise}: F_{hkl}^N = 0.$

Here f_A , f_B , f_C and f_D are average scattering lengths for the atoms at A, B, C and D sublattices, namely

$$f_\xi = [\text{Mn}]_\xi b_{\text{Mn}} + [\text{V}]_\xi b_{\text{V}} + [\text{Al}]_\xi b_{\text{Al}}, \quad (6)$$

Table 1 Possible atomic arrangements in the doubly ordered *bcc* structure illustrated in Fig. 1 for the stoichiometric composition Mn_2VAl . Atomic arrangement is represented by the occupation probability $[E]_\xi$ of E atom at ξ sublattice where $E=\text{Mn}$, V and Al, $\xi=\text{A, B, C}$ and D. The calculated reliability factor R is also given in the last column, on the assumption of stoichiometry and perfect ordering in each case.

No.	Space group	Stru.	Occupation probability, $[E]_\xi$	R
1	$\text{O}_h^5\text{-Fm}\bar{3}\text{m}$	DO_3	$[\text{Mn}]_{\text{A,B,C}}=\frac{2}{3}, [\text{V}]_{\text{A,B,C}}=\frac{1}{3}, [\text{Al}]_{\text{D}}=1$, otherwise=0	0.35
2	$\text{O}_h^5\text{-Fm}\bar{3}\text{m}$	DO_3	$[\text{Mn}]_{\text{A,B,C}}=\frac{2}{3}, [\text{V}]_{\text{D}}=1, [\text{Al}]_{\text{A,B,C}}=\frac{1}{3}$, otherwise=0	0.65
3	$\text{O}_h^5\text{-Fm}\bar{3}\text{m}$	DO_3	$[\text{Mn}]_{\text{A,B,C}}=\frac{1}{3}, [\text{Mn}]_{\text{D}}=1, [\text{V}]_{\text{A,B,C}}=\frac{1}{3}, [\text{Al}]_{\text{A,B,C}}=\frac{1}{3}$, otherwise=0	0.35
4	$\text{O}_h^5\text{-Fm}\bar{3}\text{m}$	L2_1	$[\text{Mn}]_{\text{A,C}}=1, [\text{V}]_{\text{B}}=1, [\text{Al}]_{\text{D}}=1$, otherwise=0	0.23
5	$\text{O}_h^5\text{-Fm}\bar{3}\text{m}$	L2_1	$[\text{Mn}]_{\text{A,C}}=\frac{1}{2}, [\text{Mn}]_{\text{B}}=1, [\text{V}]_{\text{A,C}}=\frac{1}{2}, [\text{Al}]_{\text{D}}=1$, otherwise=0	0.56
6	$\text{O}_h^5\text{-Fm}\bar{3}\text{m}$	L2_1	$[\text{Mn}]_{\text{A,C}}=\frac{1}{2}, [\text{Mn}]_{\text{B}}=1, [\text{V}]_{\text{D}}=1, [\text{Al}]_{\text{A,C}}=\frac{1}{2}$, otherwise=0	0.32
7	$\text{T}_d^2\text{-F}\bar{4}3\text{m}$	B32	$[\text{Mn}]_{\text{B,C}}=1, [\text{V}]_{\text{A}}=1, [\text{Al}]_{\text{D}}=1$, otherwise=0	0.61
8	$\text{O}_h^5\text{-Pm}\bar{3}\text{m}$	B2	$[\text{Mn}]_{\text{A,C}}=1, [\text{V}]_{\text{B,D}}=\frac{1}{2}, [\text{Al}]_{\text{B,D}}=\frac{1}{2}$, otherwise=0	0.40
9	$\text{O}_h^5\text{-Im}\bar{3}\text{m}$	A2	$[\text{Mn}]_{\text{A,B,C,D}}=\frac{1}{2}, [\text{V}]_{\text{A,B,C,D}}=\frac{1}{4}, [\text{Al}]_{\text{A,B,C,D}}=\frac{1}{4}$, otherwise=0	0.77

where $[\text{Mn}]_\xi$ is the occupation probability of Mn atom at ξ ($=\text{A, B, C}$ and D) sublattices and b_{Mn} the nuclear scattering amplitude⁽⁷⁾ of Mn atom, etc.

To determine the most plausible ordered structure of this alloy, we examine nine possible kinds of ordered structures indicated in Table 1 and tried to find a structure with the least value of the reliability factor R ⁽⁸⁾ defined as follows:

$$R = \sum_{hkl} ||F_{hkl}^N|_0 - |F_{hkl}^N|_c| / \sum_{hkl} |F_{hkl}^N|_0, \quad (7)$$

where $|F_{hkl}^N|_0$ is the structure factor obtained experimentally according to eq. (1) and $|F_{hkl}^N|_c$ is the one calculated from eqs. (3)–(6) in each crystal structure indicated Table 1. The occupation probability of each atom in various possible ordered structures and their reliability factors are given. For simplification, $|F_{hkl}^N|_c$ is calculated at the stoichiometric composition Mn_2VAl , though the actual composition of the alloy is slightly different from this composition.

As can be seen in Table 1, the most probable ordered structure may be No. 4 with $R=0.23$, that is, the Heusler(L2_1)-type structure. Mn atoms occupy the A and C sublattices and form the simple cubic sublattice, while V and Al atoms occupy the B and D sublattices, respectively. The structures of Nos. 1, 3 and 6 must be disregarded, though they have a lower reliability factor of 0.35 or 0.32, because in these structures the calculated intensity of the strongest peak 200 is very different from the observed one.

V. Order Parameter and Magnetic Moment

1. Order parameter to determine atomic arrangement

According to the results of the previous section, we can determine the atomic arrangement of the alloy more precisely. In an off-stoichiometric composition, the atomic arrangement of the L2_1 type ordered alloy is described generally using four long range order parameters defined as follows⁽⁹⁾:

$$\begin{aligned} S_1^* &= 2([\text{Mn}]_\alpha - x), \quad S_2^* = (4/3)([\text{V}]_\beta + x + y - 1), \\ S_3^* &= (4/3)([\text{Al}]_\gamma - y) \quad \text{and} \quad S_4^* = 2(x - [\text{Mn}]_\beta), \end{aligned} \quad (8)$$

where x and y are the atomic fraction of Mn and Al atoms in the alloy system $\text{Mn}_x\text{V}_{1-x-y}\text{Al}_y$, respectively, and $\alpha(\text{A}+\text{C})$, $\beta(\text{B})$ and $\gamma(\text{D})$ indicate the sublattices in the L2_1 structure.

Unfortunately, in neutron diffraction, the nuclear scattering amplitude does not depend on the scattering angle and therefore, only the three equations for F_{111}^N , F_{200}^N and F_{220}^N in eqs. (3)–(5) are independent. Furthermore, since F_{220}^N is determined only by the alloy composition, two equations for F_{111}^N and F_{200}^N are available to determine the order parameters. This fact forces us to reduce the number of the order parameters based on the following assumptions. Firstly, we assume that Al atom does not occupy the α sublattice site, taking into account of the result of the off-stoichiometric alloy Fe-Al ⁽¹⁰⁾, namely,

Table 2 The relation between the atomic distributions and long range order parameters in the Heusler type structure.

Elements Sites	Mn	V	Al	Total number of atoms per formula
α (A, C)	$2x + S_1^*$ $\langle 2x + S_1 \rangle$	$2(1-x-y) - S_1^*$ $-\frac{3}{4}(S_2^* - S_3^*) + \frac{1}{2}S_4^*$ $\langle 2(1-x) - S_1 \rangle$	$2y + \frac{3}{4}(S_2 - S_3^*) - \frac{1}{2}S_4^*$ $\langle 0 \rangle$	2
β (B)	$x - \frac{1}{2}S_4^*$ $\langle x - \frac{1}{2}S_1 + \frac{1}{2}S_2 - \frac{1}{2}S_3 \rangle$	$1 - x - y + \frac{3}{4}S_2^*$ $\langle 1 - x - 2y + \frac{1}{2}S_1 + \frac{1}{2}S_3 \rangle$	$y - \frac{3}{4}S_2^* + \frac{1}{2}S_4^*$ $\langle 2y - \frac{1}{2}S_2 \rangle$	1
γ (D)	$x - S_1^* + \frac{1}{2}S_4^*$ $\langle x - \frac{1}{2}S_1 - \frac{1}{2}S_2 + \frac{1}{2}S_3 \rangle$	$1 - x - y + S_1^* - \frac{3}{4}S_2^* - \frac{1}{2}S_4^*$ $\langle 1 - x - 2y + \frac{1}{2}S_1 - \frac{1}{2}S_3 \rangle$	$y + \frac{3}{4}S_2^*$ $\langle 2y + \frac{1}{2}S_2 \rangle$	1
Total number of each atom per formula	4x	4(1-x-y)	4y	$\text{Mn}_{4x}\text{V}_{4(1-x-y)}\text{Al}_{4y}$

$\langle \rangle$: Present definition.

outer: Murakami *et al.*'s definition⁽⁹⁾.

$$[\text{Al}]_\alpha = 0. \quad (9)$$

Then newly defined order parameters may be introduced as follows:

$$S_1 = S_1^* = 2([\text{Mn}]_\alpha - x),$$

$$S_2 = [\text{Al}]_\gamma - [\text{Al}]_\beta \text{ and } S_3 = [\text{V}]_\beta - [\text{V}]_\gamma. \quad (10)$$

The relation between the atomic arrangement and the three order parameters S_1 , S_2 and S_3 is given in Table 2.

2. Determination of order parameters

According to eqs. (3)–(6) and Table 2, the nuclear structure factors are obtained immediately as follows:

$$F_{111}^N = 4[(S_2 - S_3) \cdot (b_{\text{Mn}} - b_{\text{V}}) + S_2(b_{\text{V}} - b_{\text{Al}})], \quad (11)$$

$$F_{200}^N = 4[2S_1(b_{\text{Mn}} - b_{\text{V}}) + 4y(b_{\text{V}} - b_{\text{Al}})] \quad (12)$$

and

$$F_{220}^N = 4[4xb_{\text{Mn}} + 4(1-x-y)b_{\text{V}} + 4yb_{\text{Al}}], \quad (13)$$

where eq. (13) is used to obtain the coefficient K in eq. (1) so as to give the other F_{hkl}^N 's. S_1 is obtained from eq. (12). Assuming the relation of $S_2 = S_3$, S_2 is obtained easily from eq. (11). In this case Mn atoms occupy equally the B and D sublattices. The obtained results of $S_1 = 0.69$ and $S_2 = 0.85$ indicate that the annealed alloy $\text{Mn}_{0.47}\text{V}_{0.28}\text{Al}_{0.25}$ can be well ordered to the L2_1 structure. 73% of Mn atoms remain in the A and C sublattices and 85% of Al atoms in the D sublattice, because the calculated maximum

values of S_1 and S_2 are $2x = 0.94$ and $4y = 1$, respectively.

When $[\text{Al}]_\alpha > 0$ is true, S_1 may give a value larger than $2x = 0.94$ given above.

3. Magnetic moment

In the alloy $\text{Mn}_{0.47}\text{V}_{0.28}\text{Al}_{0.25}$ with L2_1 structure, Mn atom is expected to have the magnetic moment, while it is uncertain whether V atom has the magnetic moment or not. It is assumed that this alloy has three kinds of magnetic moment i.e. $\mu_{\text{Mn}}(\text{I})$, $\mu_{\text{Mn}}(\text{II})$ and $\mu_{\text{V}}(\text{II})$. Here $\mu_{\text{Mn}}(\text{I})$ denotes the magnetic moment per Mn atom at the α sublattice, and $\mu_{\text{Mn}}(\text{II})$ and $\mu_{\text{V}}(\text{II})$ denote those per Mn and V atoms at the β and γ sublattices, respectively. For simplification the magnetic moment per V atom at the α sublattice is neglected.

When these magnetic moments have a parallel alignment, the magnetic structure factors are given by

$$F_{111}^M = 1.076[(S_2 - S_3) \cdot f_{111}^{\text{Mn}^+} \cdot \mu_{\text{Mn}}(\text{II}) + S_3 f_{111}^{\text{V}} \cdot \mu_{\text{V}}(\text{II})], \quad (14)$$

$$F_{200}^M = 1.076[(2x + S_1) \cdot f_{200}^{\text{Mn}^+} \cdot \mu_{\text{Mn}}(\text{I}) - (2x - S_1) \cdot f_{200}^{\text{Mn}^+} \cdot \mu_{\text{Mn}}(\text{II}) - (2 - 2x - 4y + S_1) \cdot f_{200}^{\text{V}} \cdot \mu_{\text{V}}(\text{II})], \quad (15)$$

and

$$F_{220}^M = 1.076[(2x + S_1) \cdot f_{220}^{\text{Mn}^+} \cdot \mu_{\text{Mn}}(\text{I}) + (2x - S_1) \cdot f_{220}^{\text{Mn}^+} \cdot \mu_{\text{B}}(\text{II}) + (2 - 2x - 4y + S_1) \cdot f_{220}^{\text{V}} \cdot \mu_{\text{V}}(\text{II})], \quad (16)$$

where $f_{hkl}^{\text{Mn}+}$ and f_{hkl}^{V} are the magnetic form factors for Mn and V, respectively, and are given by Watson and Freeman⁽¹¹⁾.

If our assumptions of $[Al]_{\alpha}=0$ and $S_2=S_3$ are true, then the existence of the magnetic contribution of the 111 peak shows that $\mu_{\text{V}}(\text{II})$ has the magnetic moment of $-0.9\mu_{\text{B}}$ as shown from eq. (14) and $\mu_{\text{Mn}}(\text{I})$ and $\mu_{\text{Mn}}(\text{II})$ have $1.5\mu_{\text{B}}$ and $-1.0\mu_{\text{B}}$, respectively, as shown from eqs. (15) and (16). This result means that the annealed alloy $\text{Mn}_{0.47}\text{V}_{0.28}\text{Al}_{0.25}$ has the L2_1 structure with the equal distribution of Mn atom at the β and γ sublattices. This alloy has essentially a ferrimagnetic structure consisting of two interpenetrating simple cubic magnetic sublattices. We call it case (1).

However, there is such another possibility that $S_2 \neq S_3$, and in this case $\mu_{\text{V}}(\text{II})=0$ and the magnetic moment $\mu_{\text{Mn}}(\text{II})$ is large. We call it case (2). Results that $\mu_{\text{Mn}}(\text{I})=1.5\mu_{\text{B}}$ and $\mu_{\text{Mn}}(\text{II})=-3.4\mu_{\text{B}}$ are obtained from eqs. (15) and (16) using $S_1=0.69$. While, results that $S_2=0.66$ and $S_3=0.44$ are obtained from eqs. (11) and (14). These results mean that the annealed alloy $\text{Mn}_{0.47}\text{V}_{0.28}\text{Al}_{0.25}$ has the L2_1 structure with the unequal distribution of Mn atoms between the β and γ sublattices. This magnetic structure is essentially ferrimagnetic, and its magnetic atom is Mn only.

In Table 3(a) and 3(b), the observed intensities of the nuclear and magnetic peaks are compared with those calculated according to the above two cases. We could not determine distinctly from these results which case is more plausible.

VI. Summary and Discussions

Neutron diffraction measurements were performed on the alloy $\text{Mn}_{1.88}\text{V}_{1.12}\text{Al}_{1.00}$ with approximate composition Mn_2VAl . It was confirmed that the annealed alloy $\text{Mn}_{0.47}\text{V}_{0.28}\text{Al}_{0.25}$ has essentially the Heusler(L2_1)-type structure. In this new L2_1 structure, Mn atoms form the simple cubic sublattice α , which is very different from the ordinary Mn Heusler alloys such as Cu_2MnAl and Cu_2MnIn . Mn atoms at the sublattice α have the magnetic moment $(1.5 \pm 0.3)\mu_{\text{B}}$ per Mn atom ferromagnetically aligned. The magnetic atoms in

Table 3 Comparison of the calculated neutron diffraction intensities with the observed ones according to case (1): (a) and case (2): (b), respectively.

<i>hkl</i>	$I_{hkl}^{\text{N}} (\times 10^3 \text{ counts})$		$I_{hkl}^{\text{M}} (\times 10^3 \text{ counts})$	
	Calcu.		Calcu. (μ_{B})	
	$S_1=0.69$ $S_2=0.85$ $S_3=0.85$	Obs.	$\mu_{\text{Mn}}(\text{I})=1.5$ $\mu_{\text{Mn}}(\text{II})=-1.0$ $\mu_{\text{V}}(\text{II})=-0.9$	Obs. (μ_{B})
111	8.3	8.3 ± 0.2	1.3	1.3 ± 0.4
200	33.1	33.1 ± 0.3	11.2	11.2 ± 0.6
220	8.0	8.0 ± 0.2	1.4	1.4 ± 0.4
311	7.1	6.9 ± 0.2	0.1	—
222	14.8	11.8 ± 0.2	1.4	1.2 ± 0.4
400	2.0	2.0 ± 0.2	0.2	—
(a): case (1)				
<i>hkl</i>	$I_{hkl}^{\text{N}} (\times 10^3 \text{ counts})$		$I_{hkl}^{\text{M}} (\times 10^3 \text{ counts})$	
	Calcu.		Calcu. (μ_{B})	
	$S_1=0.69$ $S_2=0.66$ $S_3=0.44$	Obs.	$\mu_{\text{Mn}}(\text{I})=1.5$ $\mu_{\text{Mn}}(\text{II})=-3.4$ $\mu_{\text{V}}(\text{II})=0$	Obs. (μ_{B})
111	8.3	8.3 ± 0.2	1.3	1.3 ± 0.4
200	33.1	33.1 ± 0.3	11.2	11.2 ± 0.6
220	8.0	8.0 ± 0.2	1.4	1.4 ± 0.4
311	6.9	6.9 ± 0.2	0.3	—
222	14.8	11.8 ± 0.2	1.6	1.2 ± 0.4
400	2.0	2.0 ± 0.2	0.1	—
(b): case (2)				

another simple cubic sublattice composed of two f_{CC} sublattices (β and γ), may have the magnetic moments, $\mu_{\text{V}}(\text{II})=-0.9\mu_{\text{B}}$ and $\mu_{\text{Mn}}(\text{II})=-1.0\mu_{\text{B}}$, for case (1) or $\mu_{\text{V}}(\text{II})=0$ and $\mu_{\text{Mn}}(\text{II})=-3.4\mu_{\text{B}}$ for case (2). Though the magnitudes of these magnetic moments $\mu_{\text{Mn}}(\text{II})$ and $\mu_{\text{V}}(\text{II})$ may be somewhat ambiguous, one may be sure that these magnetic moments align antiparallel to the Mn moment $\mu_{\text{Mn}}(\text{I})$ in the α sublattice. Namely, there is the strong antiparallel coupling between the nearest neighbour magnetic atoms in this alloy. This fact helps us to understand the occurrence of the mictomagnetism and the thermomagnetic curve typical of ferromagnetism in the Mn-V-Al ternary system.

Based on the magnetic structure, an average magnetic moment per atom can be calculated and compared with that of the direct magnetic measurement at room temperature. The latter value is $(0.37 \pm 0.09)\mu_{\text{B}}$. This value lies around $0.38\mu_{\text{B}}$ of case (1) and $0.40\mu_{\text{B}}$ of case (2) for the

calculated values. The direct measurement of magnetization gives some complementary information about the magnetic structure of this alloy system.

Recently, NMR spin echo spectra obtained by Yoshida *et al.*⁽¹²⁾ and its analysis by Nakamichi and Stager⁽¹³⁾ suggest that V atom has the magnetic moment supporting our result of case (1). The latter work for a Mn excess alloy $\text{Mn}_{2.07}\text{V}_{1.02}\text{Al}_{0.91}$ showed also neutron diffraction patterns approximately similar to those in our study. This fact seems also to support our result of case (1).

In conclusion, the annealed alloy $\text{Mn}_{0.47}\text{V}_{0.28}\text{Al}_{0.25}$ has the Heusler type structure and the ferrimagnetic structure with the magnetic moment of $\mu_{\text{Mn}}(\text{I}) = 1.5\mu_{\text{B}}$ in the A + C(α) sublattice and those of $\mu_{\text{Mn}}(\text{II}) = -1.0\mu_{\text{B}}$ and $\mu_{\text{V}}(\text{II}) = -0.9\mu_{\text{B}}$ in the B(β) and D(γ) sublattices, respectively.

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