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# Magnetic and neutron diffraction study on quaternary oxides $M\text{TeMoO}_6$ ( $M = \text{Mn}$ and $\text{Zn}$ )

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## Abstract

Crystal structures and magnetic properties of quaternary oxides  $M\text{TeMoO}_6$  ( $M = \text{Mn}$  and  $\text{Zn}$ ) were investigated. From the Rietveld analyses for the powder x-ray and neutron diffraction measurements, their detailed structures have been determined. Both compounds have orthorhombic structure with space group  $P2_12_12$  and a charge configuration of  $M^{2+}\text{Te}^{4+}\text{Mo}^{6+}\text{O}_6$ .  $\text{ZnTeMoO}_6$  shows diamagnetic behavior. In this structure, M ions are arranged in a square-planar manner. The temperature dependence of the magnetic susceptibility for  $\text{MnTeMoO}_6$  shows a broad peak at  $\sim 33$  K, which is due to a two-dimensional characteristic of the magnetic interaction. In addition, this compound shows an antiferromagnetic transition at 20 K. The magnetic structure was determined by the powder neutron diffraction measurement at 3.3 K. The magnetic moments of  $\text{Mn}^{2+}$  ions ( $4.45 \mu_B$ ) order in a collinear antiferromagnetic arrangement along the  $b$  axis.

 Supplementary data are available from [stacks.iop.org/JPhysCM/21/046006](http://stacks.iop.org/JPhysCM/21/046006)

## 1. Introduction

Quaternary oxides  $M\text{TeMoO}_6$  ( $M = \text{Mg}, \text{Mn}, \text{Co}, \text{Zn}, \text{Cd}$ ), the so-called additive telluromolybdates, were first synthesized in the latter half of the 1970s [1–8] and their catalytic properties were investigated [1, 9–11]. The x-ray diffraction data for these compounds were indexed in an orthorhombic unit cell with space group  $P2_12_12$  [1, 12], except for  $\text{CdTeMoO}_6$  which adopts a tetragonal unit cell [8]. However, their accurate structural data was unknown for a long time. After about two decades, Hayashi *et al* proposed the atomic positions of three metal ions in  $\text{CoTeMoO}_6$  [13], and Laligant determined detailed structural parameters for the orthorhombic  $\text{CoTeMoO}_6$  and tetragonal  $\text{CdTeMoO}_6$  [14].

The schematic crystal structure of the orthorhombic  $M\text{TeMoO}_6$  is illustrated in figure 1(a). In this structure, all the M ions are located at  $z \sim 0.5$  and form a square-planar lattice in the  $ab$ -plane (see figure 1(b)); the layers consisting of  $\text{MO}_6$  octahedra are separated by other layers which are formed by  $\text{MoO}_4$  tetrahedra and  $\text{TeO}_4$  polyhedra.

If the M ion is magnetic, this compound can be seen as a two-dimensional magnetic system. The materials in such a system have been of great interest because of their unique physical properties [15]. However, their magnetic properties are not well known except for  $\text{CoTeMoO}_6$  determined by magnetic susceptibility measurements ( $T = 100\text{--}300$  K) [16], and no evidence for the long-range magnetic ordering has been observed.

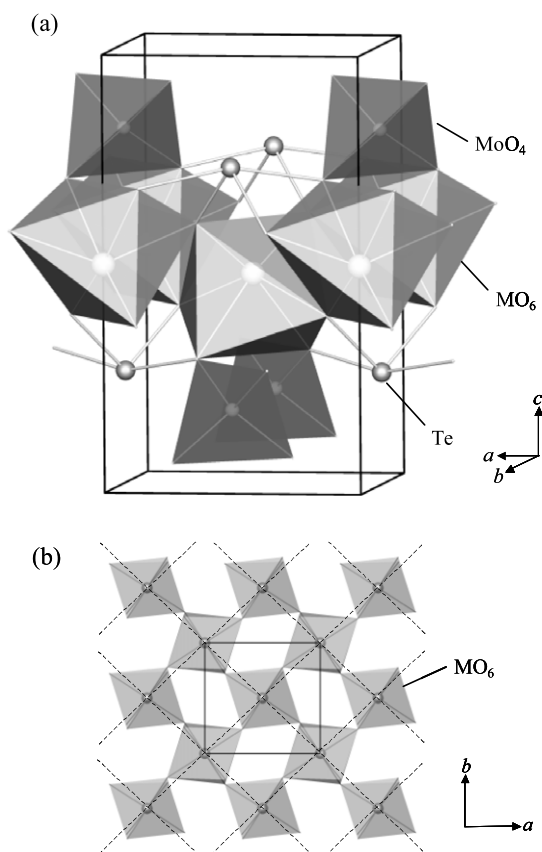
In this paper, we will report the detailed crystal structures for  $\text{MnTeMoO}_6$  and  $\text{ZnTeMoO}_6$  determined by powder x-ray and neutron diffraction measurements, and reveal the magnetic properties of  $\text{MnTeMoO}_6$  by magnetic susceptibility, specific heat, and neutron diffraction measurements at low temperatures.

## 2. Experimental details

### 2.1. Sample preparation

Two polycrystalline samples ( $\text{MnTeMoO}_6$  and  $\text{ZnTeMoO}_6$ ) were prepared by the solid-state reaction. The  $\text{ZnTeMoO}_6$  was

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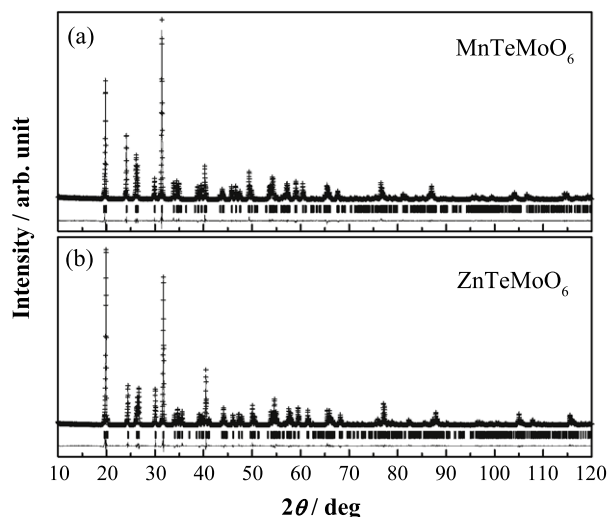


**Figure 1.** (a) Schematic crystal structure of the orthorhombic  $M\text{TeMoO}_6$  and (b) network of the  $\text{MO}_6$  octahedra. In (b), the dashed lines represent the square-planar lattice of M ions.

needed to estimate the lattice and electronic contribution to the total specific heat for  $\text{MnTeMoO}_6$ , which will be described later. The starting materials,  $\text{MnO}$ ,  $\text{ZnO}$ ,  $\text{TeO}_2$  and  $\text{MoO}_3$  were weighed in appropriate metal ratios and ground intimately in an agate mortar. The mixtures were pressed into pellets, and then the pellet was sealed in an evacuated silica tube to prevent the loss of reagents by the volatilization. These ampules were heated at 873 K (for  $\text{MnTeMoO}_6$ ) or 823 K (for  $\text{ZnTeMoO}_6$ ) for  $12 \times 3$  h with intermediate grinding and pelletizing.

### 2.2. Powder x-ray and neutron diffraction measurements

The powder x-ray diffraction (XRD) measurements were performed at room temperature in the range  $10^\circ \leq 2\theta \leq 120^\circ$  using a  $2\theta$  step size of  $0.02^\circ$  with  $\text{Cu K}\alpha$  radiation on a Rigaku MultiFlex diffractometer. Powder neutron diffraction (ND) profiles were also measured for  $\text{MnTeMoO}_6$  (at 3.3 K, 11 K and room temperature) and  $\text{ZnTeMoO}_6$  (at room temperature) in the range  $3^\circ \leq 2\theta \leq 153^\circ$  at intervals of  $0.02^\circ$  with a wavelength of  $1.81424 \text{ \AA}$ . Measurements were performed by the Kinken powder diffractometer for high efficiency and high resolution measurements, HERMES, of the Institute for Materials Research (IMR), Tohoku University [17], installed at the JRR-3M reactor in Japan Atomic Energy Agency (JAEA), Tokai. The x-ray and neutron diffraction data were analyzed by the Rietveld technique, using the program RIETAN2000 [18].



**Figure 2.** Powder x-ray diffraction profiles for (a)  $\text{MnTeMoO}_6$  and (b)  $\text{ZnTeMoO}_6$ . The calculated and observed diffraction profiles are shown on the top as a solid line and cross markers, respectively. The vertical markers show positions calculated from Bragg reflections. The bottom trace is a plot of the difference between the calculated and observed intensities.

### 2.3. Magnetic susceptibility and specific heat measurements

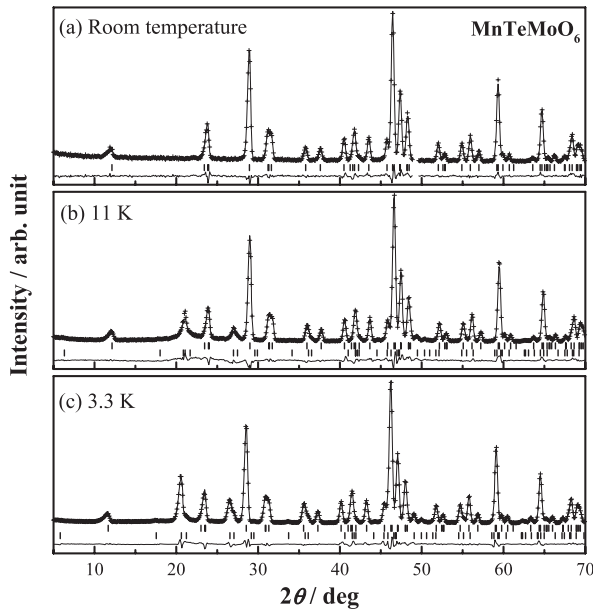
The temperature dependence of the magnetic susceptibilities was measured under both zero-field-cooled (ZFC) and field-cooled (FC) conditions in an applied field of 0.1 T over the temperature range 1.8–400 K using a SQUID magnetometer (Quantum Design, MPMS-5S). In addition, specific heat measurements were performed using a relaxation technique with a commercial physical property measurement system (Quantum Design, PPMS model) in the temperature range 1.8–300 K. The sintered sample in the form of a pellet was mounted on a thin alumina plate with grease for better thermal contact.

## 3. Results and discussion

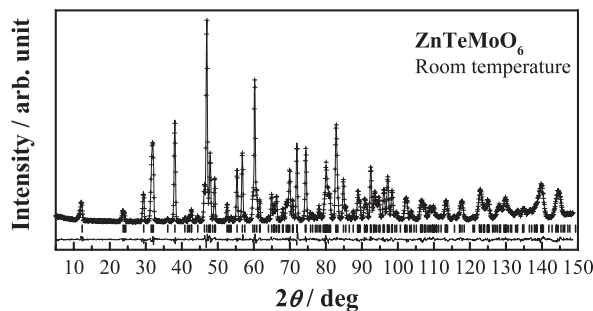
### 3.1. Crystal structure

The title compounds  $\text{MnTeMoO}_6$  and  $\text{ZnTeMoO}_6$  were successfully prepared as a single phase. Figure 2 shows their powder x-ray diffraction profiles. Both data were indexed with an orthorhombic unit cell ( $a \sim 5.3 \text{ \AA}$ ,  $b \sim 5.0 \text{ \AA}$ , and  $c \sim 8.9 \text{ \AA}$ ) with the space group  $P2_12_12$  (No. 18), and analyzed by the Rietveld method using the structural model for  $\text{CoTeMoO}_6$  [14]. The calculated profiles are plotted in figure 2, which gives a good agreement with the observed profiles. The obtained lattice parameters are consistent with those in earlier works [1, 12]. The structural parameters determined by XRD measurements are shown in supplementary tables (available at [stacks.iop.org/JPhysCM/21/046006](http://stacks.iop.org/JPhysCM/21/046006)).

The powder neutron diffraction measurements were performed for  $\text{MnTeMoO}_6$  (at 3.3 K, 11 K, and room temperature) and  $\text{ZnTeMoO}_6$  (at room temperature). Their diffraction profiles are shown in figures 3 and 4, respectively. The Rietveld analyses of the data indicate that both compounds adopt the same structure as  $\text{CoTeMoO}_6$  [14], and are consistent



**Figure 3.** Powder neutron diffraction profiles for MnTeMoO<sub>6</sub> (a) at room temperature, (b) at 11 K, and (c) at 3.3 K. The calculated and observed diffraction profiles are shown on the top as a solid line and cross markers, respectively. The upper vertical markers show positions calculated from nuclear Bragg reflections, and in (b) and (c) the lower ones show those from magnetic Bragg reflections. The bottom trace is a plot of the difference between the calculated and observed intensities.



**Figure 4.** Powder neutron diffraction profile for ZnTeMoO<sub>6</sub> at room temperature. The calculated and observed diffraction profiles are shown on the top as a solid line and cross markers, respectively. The vertical markers show positions calculated from nuclear Bragg reflections. The bottom trace is a plot of the difference between the calculated and observed intensities.

with the results of the XRD data. Evidence of the occurrence of cation disorder or an oxygen defect has not been found; all the occupancy parameters were fixed to 1.0. For MnTeMoO<sub>6</sub>, the data collected at 3.3 and 11 K show a number of low-angle peaks, which are not observed at room temperature. As will be discussed later, they are due to an antiferromagnetic ordering of Mn<sup>2+</sup> moments. The crystal structures at 3.3 and 11 K have the same symmetry as that at room temperature; thus, no structural transition occurs. The refined structural parameters for MnTeMoO<sub>6</sub> and ZnTeMoO<sub>6</sub> are summarized in tables 1 and 2, respectively.

The crystal structure of the orthorhombic MTeMoO<sub>6</sub> is illustrated in figure 1(a). In this structure, M ions form a

**Table 1.** Structural parameters for MnTeMoO<sub>6</sub> determined by powder neutron diffraction measurements.

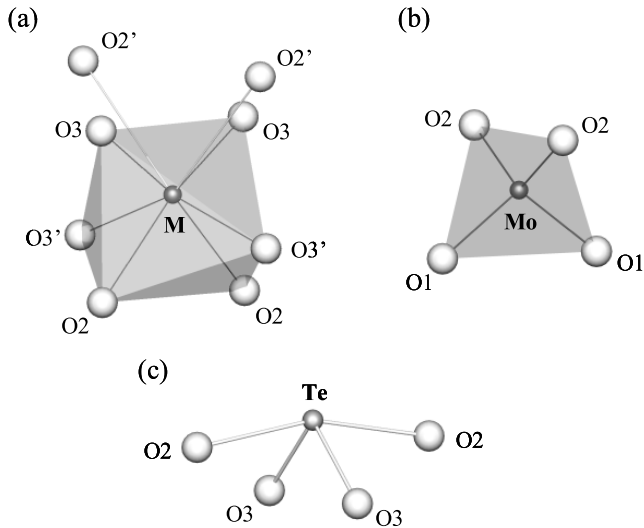
Atom	Site	<i>x</i>	<i>y</i>	<i>z</i>	<i>B</i> (Å <sup>2</sup> )
Room temperature, space group <i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2; <i>a</i> = 5.2941(3) Å, <i>b</i> = 5.1350(3) Å, <i>c</i> = 8.9544(5) Å, <i>R</i> <sub>wp</sub> = 6.45%, <i>R</i> <sub>p</sub> = 5.06%, <i>R</i> <sub>e</sub> = 4.75%, <i>R</i> <sub>I</sub> = 1.01%.					
Mn	2 <i>a</i>	0	0	0.5229(7)	0.89(9)
Te	2 <i>b</i>	0	1/2	0.2497(5)	0.59(9)
Mo	2 <i>b</i>	0	1/2	0.8102(4)	0.52(8)
O1	4 <i>c</i>	0.2986(5)	0.8391(5)	0.0722(3)	1.02(7)
O2	4 <i>c</i>	0.1895(5)	0.7478(6)	0.7086(3)	0.77(7)
O3	4 <i>c</i>	0.1773(6)	0.7108(5)	0.3867(3)	0.90(7)
<i>T</i> = 11 K, space group <i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2; <i>a</i> = 5.2850(3) Å, <i>b</i> = 5.1198(2) Å, <i>c</i> = 8.9132(4) Å, <i>R</i> <sub>wp</sub> = 7.40%, <i>R</i> <sub>p</sub> = 5.40%, <i>R</i> <sub>e</sub> = 2.77%, <i>R</i> <sub>I</sub> (crystal) = 1.56%, <i>R</i> <sub>I</sub> (magnetic) = 1.53%, μ <sub>Mn</sub> = 2.71(2)μ <sub>B</sub> .					
Mn	2 <i>a</i>	0	0	0.5227(6)	0.14(9)
Te	2 <i>b</i>	0	1/2	0.2486(4)	0.29(8)
Mo	2 <i>b</i>	0	1/2	0.8111(4)	0.22(7)
O1	4 <i>c</i>	0.2972(5)	0.8394(5)	0.0712(3)	0.44(5)
O2	4 <i>c</i>	0.1877(5)	0.7500(5)	0.7093(3)	0.26(5)
O3	4 <i>c</i>	0.1781(5)	0.7133(4)	0.3862(3)	0.43(5)
<i>T</i> = 3.3 K, space group <i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2; <i>a</i> = 5.2846(2) Å, <i>b</i> = 5.1195(2) Å, <i>c</i> = 8.9119(4) Å, <i>R</i> <sub>wp</sub> = 5.34%, <i>R</i> <sub>p</sub> = 3.82%, <i>R</i> <sub>e</sub> = 1.82%, <i>R</i> <sub>I</sub> (crystal) = 0.95%, <i>R</i> <sub>I</sub> (magnetic) = 1.98%, μ <sub>Mn</sub> = 4.46(2)μ <sub>B</sub> .					
Mn	2 <i>a</i>	0	0	0.5227(3)	0.09(9)
Te	2 <i>b</i>	0	1/2	0.2483(3)	0.21(6)
Mo	2 <i>b</i>	0	1/2	0.8120(3)	0.20(6)
O1	4 <i>c</i>	0.2970(4)	0.8400(4)	0.0710(2)	0.45(5)
O2	4 <i>c</i>	0.1879(4)	0.7492(4)	0.7086(2)	0.37(5)
O3	4 <i>c</i>	0.1775(4)	0.7128(3)	0.3863(2)	0.40(5)

**Table 2.** Structural parameters for ZnTeMoO<sub>6</sub> determined by powder neutron diffraction measurements at room temperature. (Note: space group *P*2<sub>1</sub>2<sub>1</sub>2; *a* = 5.2630(2) Å, *b* = 5.0426(3) Å, *c* = 8.9105(4) Å, *R*<sub>wp</sub> = 5.60%, *R*<sub>p</sub> = 4.29%, *R*<sub>e</sub> = 4.04%, *R*<sub>I</sub> = 0.90%.)

Atom	Site	<i>x</i>	<i>y</i>	<i>z</i>	<i>B</i> (Å <sup>2</sup> )
Zn	2 <i>a</i>	0	0	0.5218(4)	0.46(7)
Te	2 <i>b</i>	0	1/2	0.2553(3)	0.56(7)
Mo	2 <i>b</i>	0	1/2	0.8088(3)	0.46(7)
O1	4 <i>c</i>	0.2891(4)	0.8467(4)	0.0745(3)	1.13(6)
O2	4 <i>c</i>	0.1805(5)	0.7612(4)	0.7067(2)	0.77(6)
O3	4 <i>c</i>	0.1746(4)	0.7213(4)	0.3933(2)	0.66(6)

square-planar lattice with 3.71 Å on a side for M = Mn and 3.66 Å for Zn (see figure 1(b)). This lattice is very close to a regular one but somewhat uneven (±0.2 Å from the horizontal level) because M ions locate in the 2*a* site: (0, 0, *z*) and (1/2, 1/2, 1−*z*) with *z* ~ 0.52. The distance between adjacent layers of M ions (*c* = 8.91–8.95 Å) is much longer than the distance of the nearest M–M. Therefore, MnTeMoO<sub>6</sub> is appropriate to observe a two-dimensional magnetic behavior.

The coordination around the M ion is shown in figure 5(a), and interatomic distances are listed in table 3. The M ion is surrounded by eight oxygen ions; among them, six oxygen ions with shorter M–O lengths (2.0–2.3 Å) form a distorted MO<sub>6</sub> octahedron, and the other two have longer M–O lengths



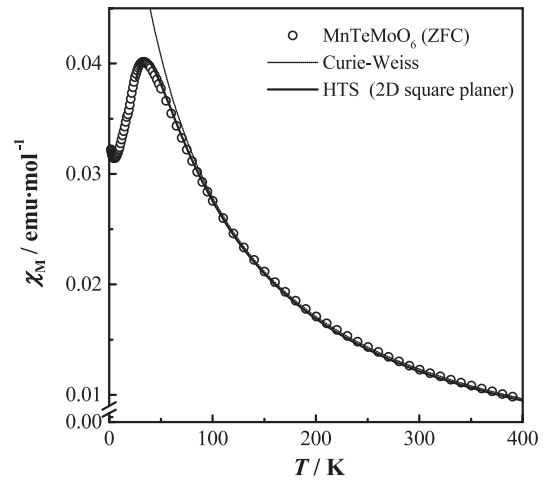
**Figure 5.** Coordination environments around (a) M, (b) Mo, and (c) Te ions in the orthorhombic MTeMoO<sub>6</sub>.

**Table 3.** Selected interatomic distances (Å) and bond valence sums determined by neutron diffraction measurements.

	ZnTeMoO <sub>6</sub> RT	MnTeMoO <sub>6</sub> RT	MnTeMoO <sub>6</sub> 3.3 K
M–O3 × 2	2.033(3)	2.139(5)	2.126(3)
M–O3' × 2	2.180(2)	2.179(4)	2.179(2)
M–O2 × 2	2.251(3)	2.334(5)	2.319(3)
M–O2' × 2	2.951(3)	2.935(5)	2.932(3)
BVS	1.90	2.03	2.08
Te–O3 × 2	1.897(3)	1.886(3)	1.892(3)
Te–O2 × 2	2.096(3)	2.126(3)	2.125(2)
Te–O1 × 2	2.823(3)	2.839(3)	2.827(2)
Te–O1' × 2	3.297(4)	3.184(5)	3.150(3)
BVS	4.19	4.17	4.14
Mo–O1 × 2	1.706(3)	1.711(4)	1.705(3)
Mo–O2 × 2	1.861(3)	1.859(3)	1.861(2)
Mo–O3 × 2	2.855(3)	2.869(4)	2.862(3)
Mo–O1' × 2	3.313(3)	3.322(3)	3.290(2)
BVS	5.90	5.87	5.92

(~2.9 Å). The bond valence sums (BVSs) calculated from the determined interatomic distances are 2.03 for M = Mn and 1.90 for Zn, which are reasonable values for divalent M ions.

The coordination environments around Mo and Te ions are illustrated in figures 5(b) and (c), respectively. The metal–oxygen distance and BVS are also listed in table 3. The values of BVS are reasonable to the hexavalent Mo and tetravalent Te ions. The interatomic distances indicate that both ions occupy four-coordinated sites; however, the way of the coordination is considerably different between them. The Mo<sup>6+</sup> ion is surrounded by four oxygen ions with a normal tetrahedral arrangement, while the Te<sup>4+</sup> ion protrudes from a polyhedron consisting of four oxygen ions, and the upper side of the Te ion is unoccupied as shown in figure 5(c). The latter is due to the existence of lone pair electrons. Similar coordination environments are often observed for Te<sup>4+</sup> compounds, e.g. CoTeMoO<sub>6</sub> [14], Co<sub>6</sub>Te<sub>5</sub>O<sub>16</sub> [19], and Ln<sub>2</sub>Te<sub>4</sub>O<sub>11</sub> (Ln = lanthanides) [20].



**Figure 6.** Temperature dependence of the ZFC magnetic susceptibility for MnTeMoO<sub>6</sub>. The dotted and solid curves represent the calculated susceptibilities by using the Curie–Weiss law and high temperature series (HTS) expansion for a square-planar lattice, respectively.

### 3.2. Magnetic susceptibility

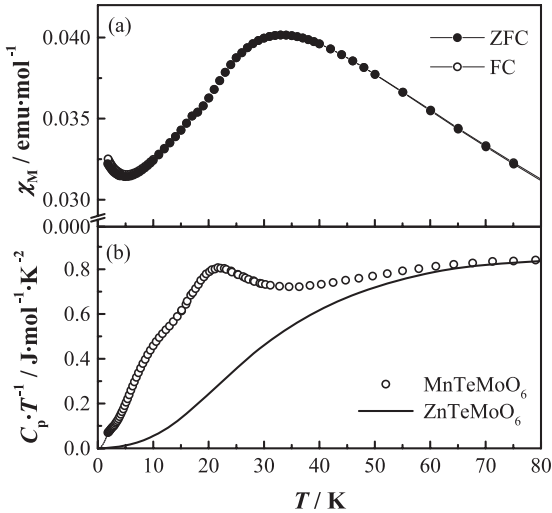
The magnetic susceptibility of ZnTeMoO<sub>6</sub> is diamagnetic and temperature-independent ( $\sim -1.3 \times 10^{-4}$  emu mol<sup>-1</sup>) in the experimental temperature range (1.8–400 K). This behavior corresponds to experimental results in the structural analysis: the valence configuration Zn<sup>2+</sup>Te<sup>4+</sup>Mo<sup>6+</sup>O<sub>6</sub> and no oxygen defect.

Figure 6 shows the temperature dependence of the ZFC magnetic susceptibility for MnTeMoO<sub>6</sub>. The data in the higher temperature region ( $T > 100$  K) were fitted by the Curie–Weiss law; the effective magnetic moment ( $\mu_{\text{eff}}$ ) and Weiss constant ( $\theta$ ) were determined to be 5.95(1)  $\mu_B$  and  $-58.8(6)$  K, respectively. This magnetic moment is in good agreement with the free-ion value (5.92  $\mu_B$ ) of the Mn<sup>2+</sup> ion in the high-spin state ( $S = 5/2$ ). The negative Weiss constant suggests that the predominant magnetic interaction between Mn ions is antiferromagnetic.

At lower temperatures, the deviation from the Curie–Weiss curve becomes larger and the magnetic susceptibility shows a broad maximum at around 33 K. This behavior may be due to the two-dimensional nature of the magnetic interaction. In this compound, the arrangement of the magnetic Mn<sup>2+</sup> ions is regarded as the square-planar lattice; thus, we have fitted the observed magnetic susceptibility using a high temperature series (HTS) expansion of the square-planar lattice for the Heisenberg model:

$$\chi_M = \frac{N_A g^2 \mu_B^2 S(S+1)}{3k_B T} \sum a_n (J/k_B T)^n \quad (1)$$

where  $N_A$ ,  $g$ ,  $\mu_B$ ,  $k_B$ , and  $J$  are the Avogadro number,  $g$  factor, Bohr magneton, Boltzmann constant, and exchange integral, respectively. We used the coefficients  $a_n$  ( $n = 1-8$ ) given in previous results [15, 21–24]. The fitting equation was obtained by applying the Padé approximation ([4,4] Padé) to the equation (1). The calculated susceptibility is shown



**Figure 7.** Temperature dependence of (a) the ZFC and FC magnetic susceptibilities for MnTeMo<sub>6</sub> and (b) specific heat divided by temperature for MnTeMo<sub>6</sub> and ZnTeMo<sub>6</sub>.

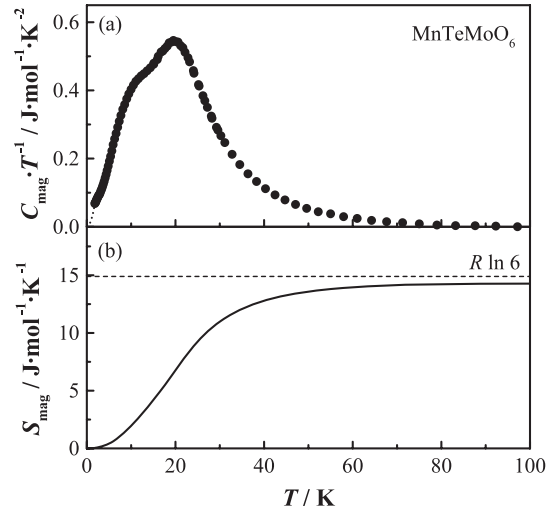
as a solid line in figure 6, and it is in good agreement with the experimental data. The  $g$  factor of the Mn<sup>2+</sup> ion and exchange integral  $J$  for the nearest Mn–Mn in the square-planar lattice are determined to be 1.95(1) and  $-1.96(1)$  K, respectively. The Weiss constant calculated from only this  $J$  value ( $\sum_i 2z_i J_i S(S+1)/3k_B$ ;  $z = 4$ ) is  $-45.7$  K, which is comparable to the value estimated from the Curie–Weiss law ( $-58.8$  K).

The ZFC and FC magnetic susceptibilities for MnTeMo<sub>6</sub> at low temperatures are plotted in figure 7(a). In addition to the broad peak at 33 K, they show a small anomaly at  $\sim 17$  K, and a slight divergence between ZFC and FC data is observed below this temperature, indicating the existence of a small ferromagnetic moment  $\sim 10^{-4} \mu_B$ . In order to obtain further information about the magnetic properties of this compound, specific heat measurements were carried out.

### 3.3. Specific heat

The temperature dependence of the specific heat divided by temperature ( $C_p/T$ ) for MnTeMo<sub>6</sub> is plotted in figure 7(b). The data show a broad  $\lambda$ -type anomaly at 20 K, which indicates that long-range antiferromagnetic ordering of Mn<sup>2+</sup> ions occurs. This result corresponds to the divergence between ZFC and FC magnetic susceptibilities. A small ferromagnetic moment observed in the magnetization measurement may be due to a weak ferromagnetic component associated with the antiferromagnetism caused by the Dzyaloshinsky–Moriya (DM) interaction, which is often observed in compounds with lower crystal symmetry.

The magnetic entropy change ( $\Delta S_{\text{mag}}$ ) associated with the antiferromagnetic transition was calculated by using  $\Delta S_{\text{mag}} = \int (C_{\text{mag}}/T)dT$ , in which the magnetic specific heat ( $C_{\text{mag}}$ ) was estimated by subtracting the lattice and electronic specific heat from the experimental specific heat of MnTeMo<sub>6</sub>. For the lattice and electronic contributions, we used the specific heat of a nonmagnetic and isostructural compound ZnTeMo<sub>6</sub>



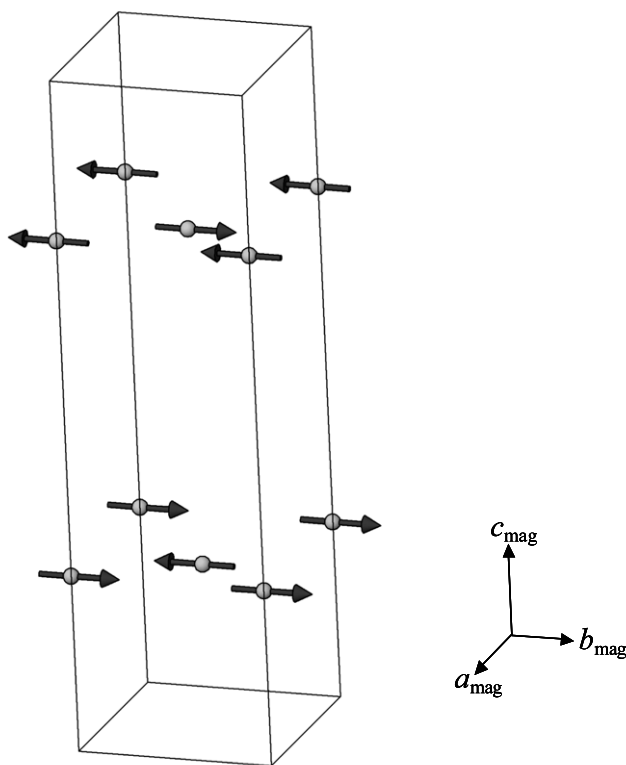
**Figure 8.** Temperature dependence of (a) the magnetic specific heat divided by temperature and (b) magnetic entropy for MnTeMo<sub>6</sub>.

(the data are shown as a solid line in figure 7(b)). The temperature dependences of the magnetic specific heat divided by temperature ( $C_{\text{mag}}/T$ ) and magnetic entropy ( $S_{\text{mag}}$ ) are plotted in figures 8(a) and (b), respectively. Above 20 K, the peak of  $C_{\text{mag}}/T$  data shows a long tail up to  $\sim 70$  K. This result indicates that the short-range magnetic ordering in the 2D layer of Mn<sup>2+</sup> ions begins at a much higher temperature than the transition temperature. The  $\Delta S_{\text{mag}}$  associated with the antiferromagnetic transition reaches  $14.3 \text{ J mol}^{-1} \text{ K}^{-1}$  at 100 K, which is in good agreement with the value expected from the sixfold-degenerated ground state of the high-spin 3d<sup>5</sup> ion ( $R \ln 6 = 14.90 \text{ J mol}^{-1} \text{ K}^{-1}$ ).

### 3.4. Magnetic structure

Powder neutron diffraction measurements for MnTeMo<sub>6</sub> were carried out below the magnetic transition temperature. The ND profiles collected at 11 and 3.3 K are plotted in figures 3(b) and (c), respectively. Both data show a number of low-angle peaks, which were associated with the antiferromagnetic transition found in the magnetic susceptibility and specific heat measurements. All these reflections can be indexed using a propagation vector  $\mathbf{k} = (0, 0, 1/2)$ , i.e. the magnetic unit cell is represented as  $a_{\text{mag}} = a$ ,  $b_{\text{mag}} = b$ , and  $c_{\text{mag}} = 2c$ . To determine the magnetic structure, we have tested many models expected from the symmetry of the crystal structure and the propagation vector, and finally found that a collinear antiferromagnetic structure gave a good calculation result. The calculated profiles are plotted as the solid line in figures 3(b) and (c). The ferromagnetic component expected from the result of the magnetic susceptibility measurement is too small to be detected.

The magnetic structure determined for MnTeMo<sub>6</sub> is illustrated in figure 9. All the magnetic moments of Mn ions are collinear along the  $b$  axis. In the  $ab$  plane, the nearest Mn moments order antiferromagnetically via the superexchange Mn–O3–Mn or longer Mn–O2–Mn pathways. The ordered



**Figure 9.** Magnetic structure of  $\text{MnTeMoO}_6$ . Diamagnetic ions are omitted. Arrows represent the direction of magnetic moments for  $\text{Mn}^{2+}$  ions.

magnetic moment for the  $\text{Mn}^{2+}$  ion is determined to be  $4.45(1) \mu_B$  at 3.3 K, which is in good agreement with the  $5 \mu_B$  expected from the  $3d^5$  electronic configuration in the high-spin state ( $S = 5/2$ ).

#### 4. Summary

Quaternary compounds  $\text{MnTeMoO}_6$  and  $\text{ZnTeMoO}_6$  adopt the orthorhombic structure with space group  $P2_12_12$ , and their detailed structural parameters have been determined by Rietveld analyses for powder x-ray and neutron diffraction measurements. The  $\text{MnTeMoO}_6$  shows characteristic magnetic properties as a two-dimensional magnet reflecting the arrangement of Mn ions (square-planar lattice). In addition,

this compound shows a long-range antiferromagnetic ordering of  $\text{Mn}^{2+}$  moments at 20 K with a collinear antiferromagnetic arrangement along the  $b$  axis.

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