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Comparative studies of the thermal conductivity of spinel oxides with orbital degrees of freedom

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We studied the thermal conductivity of various transition-metal oxides with the spinel structure (MnV_2O_4, FeV_2O_4, CoV_2O_4, and Mn_3O_4) upon varying the temperature and magnetic field. We found that for the spinel oxides having V^{3+} ions (two electrons in the t_{2g} states) at the octahedral site, the orbital ordering suppresses the thermal resistivity (inverse thermal conductivity) in contrast to purely magnetic ordering, indicating that the orbital fluctuation of the V t_{2g} states is the main factor affecting the thermal conductivity. On the other hand, for Mn_3O_4, which has Mn^{3+} ions (one electron in the e_g states) at the octahedral site, the thermal resistivity is suppressed in association with the successive magnetic phase transitions with decreasing temperature and increasing magnetic field. This can be explained by the frustration of Mn^{3+} spins and the fluctuation of Mn^{3+} e_g orbitals.

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I. INTRODUCTION

It has been shown that when heat is carried by acoustic phonons in crystals, their thermal transport properties are often dominated by the fluctuation of some degrees of freedom, which scatters acoustic phonons [1–5]. For example, the thermal conductivity of perovskite manganites exhibiting colossal magnetoresistance sharply increases below the ferromagnetic transition temperature [1]. It was proposed that such a change in the thermal conductivity is caused by a transition from an amorphous-like disordered lattice at high temperatures, by which acoustic phonons are heavily scattered, to a crystalline lattice at low temperatures, and such a variation in the lattice is likely to be associated with an order-disorder transition of the orbital degrees of freedom for Mn^{3+} ions ($3d^4$, one electron in the e_g state). Another example is observed in ferroelectric hexagonal RMnO$_3$ (R is rare earth), in which Mn^{3+} ions form a triangular lattice. The thermal conductivity of this series of compounds sharply increases below the antiferromagnetic ordering temperature (T_N) of Mn^{3+} spins [2]. In this series of compounds, strong spin fluctuation exists above T_N, originating from the geometrical frustration in the spin configuration, but it is suppressed below T_N. Since acoustic phonons are scattered by such spin fluctuation, the variation in the spin fluctuation results in the variation in the thermal conductivity. In both the above cases, the fluctuation of some degrees of freedom (orbital or spin) acts as a source of scattering for acoustic phonons by which heat is carried, and an increase in the fluctuation causes a decrease in the thermal conductivity. This is different from the situation in some compounds where heat is carried by the excitation of some degrees of freedom (magnons, for example) and accordingly, the fluctuation enhances the thermal conductivity [6,7].

Spinel oxides including transition metals are a series of compounds that may exhibit both spin and orbital fluctuations. For example, in spinel MnV_2O_4, ferrimagnetic ordering and the orbital ordering of the triply degenerate t_{2g} states of V^{3+} ions ($3d^2$), which is accompanied by a structural phase transition from cubic to tetragonal, simultaneously occur at $T_c = 57$ K [8]. In the ferrimagnetic phase, all the Mn^{2+} ($3d^5$) spins are aligned with the c axis of the tetragonal phase, whereas V^{3+} spins are canted from the c axis, though the sum of all the V spins is along the c axis, opposite to the direction of the Mn^{2+} spins [9]. The thermal resistivity (inverse thermal conductivity) of MnV_2O_4 markedly decreases below T_c [10] and exhibits a sharp change upon the application of a magnetic field near T_c [11]. The magnitude of the fluctuation of orbitals coupled with spins has been calculated by a mean-field approximation and found to scale with the thermal resistivity observed experimentally upon varying both the temperature and the magnetic field [11]. It has also been found that upon Al doping into the V site of MnV_2O_4, the orbital ordering and structural phase transition disappear and the decrease in the thermal resistivity also disappears, whereas the ferrimagnetic ordering remains almost unchanged [11]. This result supports the idea that the thermal conduction of this series of compounds is dominated by orbital fluctuation rather than by spin fluctuation.

The purpose of the present study is to investigate the thermal transport properties of the related compounds of MnV_2O_4, FeV_2O_4 [12–20], CoV_2O_4 [21–24], and Mn_3O_4 [25–34], and clarify what determines the temperature and magnetic-field dependence of the thermal conductivity in strongly correlated electron systems having multiple degrees of freedom.

FeV_2O_4 and CoV_2O_4 are similar to MnV_2O_4 in that there are V^{3+} ions at the B site (octahedral site) of the spinel structure and the A site (tetrahedral site) is occupied by magnetic divalent ions. In FeV_2O_4, there are successive structural phase transitions from cubic to tetragonal with $c < a$ at $T_1 = 140$ K, from tetragonal to orthorhombic at $T_2 = 105$ K, and from orthorhombic to tetragonal with $c > a$ at $T_3 = 70$ K. Ferrimagnetic ordering occurs at the second structural phase transition ($T_N = T_2 = 105$ K). Such successive phase transitions are caused not only by V^{3+} ions at the B site but also by orbital-active Fe^{2+} ($3d^6$) ions at the A site, in which there are three electrons in the e state. On the other hand, in CoV_2O_4, where the A site is occupied by Co^{2+} ions ($3d^7$) without any orbital degrees of freedom, ferrimagnetic ordering occurs at $T_N \sim 170$ K, but no structural phase transition exists down to 4 K.

Mn_3O_4 is similar to MnV_2O_4 in that there are Mn^{2+} ($3d^5$) ions at the A site of the spinel structure, while the B site is occupied by Mn^{3+} ($3d^4$) ions. Since Mn^{3+} ions at the octahedral site, where there is only one electron in the doubly degenerate e_g states, are subject to the strong instability of the Jahn-Teller distortion, Mn_3O_4 exhibits a structural phase transition from cubic to tetragonal at 1443 K; accordingly, the orbital degrees of freedom of Mn^{3+} ions are already quenched at room temperature. Mn_3O_4 exhibits a ferrimagnetic ordering at $T_N = 41$ K, where the spin configuration is the so-called Yafet-Kittel type, which is similar to the spin structure of MnV_2O_4 but the moments are oriented not along the [001] direction but along the [110] direction. Mn_3O_4 exhibits successive magnetic phase transitions from the Yafet-Kittel phase to an incommensurate phase at $T_1 = 39$ K and from the incommensurate phase to the so-called "cell-doubled" phase at $T_2 = 33$ K, where the magnetic unit cell is doubled along the [110] direction from that of the original cubic spinel structure. Note that it was theoretically shown that various magnetic structures exist in spinel oxides but the Yafet-Kittel phase exists only in the presence of tetragonal distortion [35,36].

In this paper, we report the thermal conductivity of these compounds upon varying the temperature and magnetic field, and how the thermal conductivity changes with various phase transitions.

II. EXPERIMENT

Single crystals of FeV_2O_4, CoV_2O_4, and Mn_3O_4 were grown by the floating-zone technique. For a further comparative study, a single crystal of $CoAl_2O_4$, which has no transition metal at the *B* site of the spinel structure, was grown by the same technique. The orientation of the crystals was determined by the Laue method. Thermal conductivity was measured by a one-heater, two-thermometer technique at temperatures from 200 K to 20 K and in magnetic fields from −5 T to 5 T. The sample was attached to a heat bath using conducting epoxy cured at 100 °C to prevent it from becoming detached from the heat bath by the magnetic force. In the following, [100] corresponds to the direction along the principal axis of the cubic spinel structure, which is parallel to the V(Mn)-O direction of $V(Mn)O_6$ octahedra.

III. RESULTS

Figure 1 shows the temperature (T) dependence of the thermal resistivity (ρ) for the four samples and MnV_2O_4 (Ref. [11]). The three AV_2O_4 samples ($A = $ Mn, Fe, Co) exhibit a similar T dependence of the thermal resistivity ρ above ∼70 K; the absolute value of ρ is ∼0.3 mK/W (the thermal conductivity is ∼3 W/mK) at 200 K, and gradually increases with decreasing T. The thermal resistivity ρ of MnV_2O_4 sharply decreases below T_c, the transition temperature of the simultaneous ferrimagnetic and orbital ordering, as discussed above.

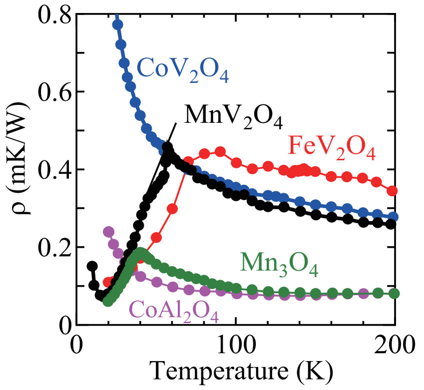


FIG. 1. (Color online) Temperature dependence of the thermal resistivity (inverse thermal conductivity) for five spinel oxides containing transition metals.

As shown in Fig. 1 and Fig. 2(a), the thermal resistivity ρ of FeV_2O_4 decreases at ∼70 K, which corresponds to T_3, the structural phase transition from orthorhombic to low-T tetragonal. This result suggests that this structural phase transition is accompanied by an orbital ordering of the V t_{2g} states. Recently, it has been shown that the magnetic structure changes from a collinear ferrimagnetic structure above T_3 to a noncollinear magnetic structure below T_3 [14]. Furthermore, the noncollinear magnetic structure of FeV_2O_4 is different from that of MnV_2O_4 in that the direction of V spins in FeV_2O_4 when projected onto the xy plane is 45° different from that in MnV_2O_4; thus, V spins are oriented in the $\langle 111 \rangle$ direction of VO_6 octahedra. This indicates that in FeV_2O_4, the local trigonal distortion of VO_6 octahedra existing in the spinel structure induces a splitting of the V t_{2g} states into a nondegenerate a_g state with a lower energy and doubly degenerate e_g states, and the degeneracy of the e_g states is lifted by spin-orbit coupling [14]. The result for the thermal resistivity in FeV_2O_4 can be interpreted in such a way that orbital ordering of the V t_{2g} states dominated by single-site spin-orbit coupling occurs at T_3, which suppresses the orbital fluctuation of the V t_{2g} states and the thermal resistivity as a consequence.

On the other hand, the thermal resistivity ρ of CoV_2O_4 increases monotonically with decreasing T, with no anomaly even at $T_N = 170$ K, down to the lowest T [Fig. 3(a)]. This behavior is similar to that of $MnV_{1.8}Al_{0.2}O_4$, in which there is a ferrimagnetic ordering but orbital ordering does not occur because of the disorder effect induced by Al substation for the V site [11]. The present result is consistent with the idea that

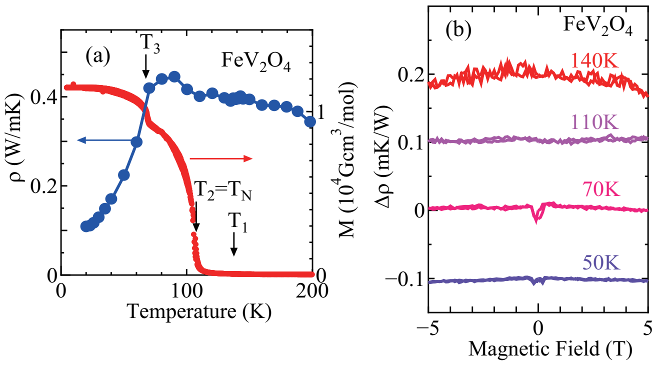


FIG. 2. (Color online) (a) Temperature dependence of the thermal resistivity and magnetization at 0.1 T for FeV_2O_4. (b) Magnetic-field dependence of the thermal resistivity at various temperatures for FeV_2O_4. The data at different temperatures are offset for clarity.

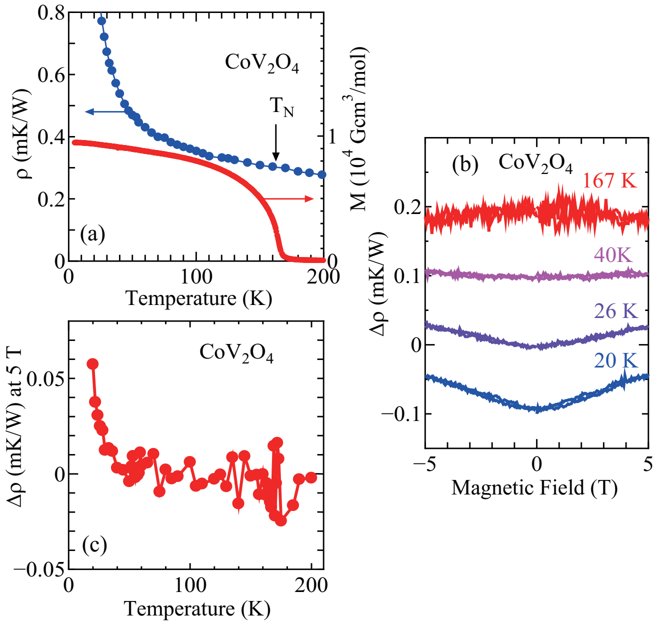


FIG. 3. (Color online) (a) Temperature dependence of the thermal resistivity and magnetization at 0.1 T for CoV_2O_4. (b) Magnetic-field dependence of the thermal resistivity at various temperatures for CoV_2O_4. The data are offset for clarity. (c) Temperature dependence of the change in the thermal resistivity with an applied magnetic field of 5 T for CoV_2O_4.

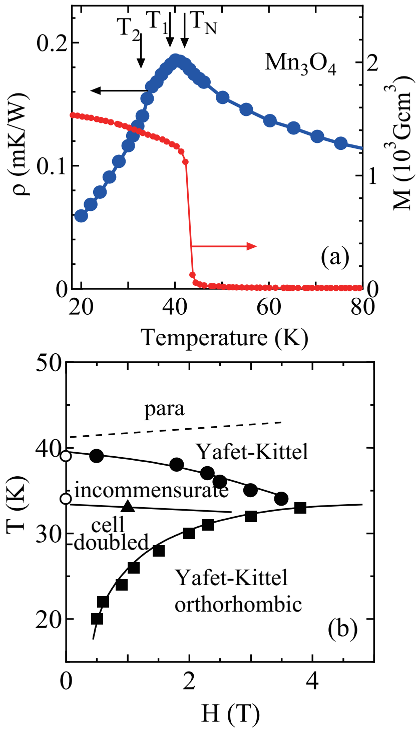


FIG. 4. (Color online) (a) Temperature dependence of the thermal resistivity and magnetization at 0.01 T for Mn_3O_4. (b) Positions of the anomalies in the magnetic-field dependence of the thermal resistivity for Mn_3O_4 shown in Fig. 5 in the space of temperature (T) and magnetic field (H). The solid and dashed lines are the phase boundaries reported in Ref. [32].

the thermal resistivity is heavily suppressed by orbital ordering but barely changes at a purely magnetic phase transition in this series of compounds.

Compared with AV_2O_4, the absolute value of ρ for Mn_3O_4, which is measured along the [100] direction in the ab plane, is much smaller ($\sim$0.1 mK/W at 200 K), which is comparable to that of $CoAl_2O_4$, which has no transition metal at the B site (Fig. 1). This result is consistent with the idea that the orbital degrees of freedom of Mn^{3+} ions at the B site are already quenched at room temperature because of the tetragonal distortion occurring at high T. The thermal resistivity increases with decreasing T down to T_N, but below T_N, it decreases with decreasing T, as shown in Fig. 4(a). In neutron diffraction measurement [27], diffuse scattering appears above T_N, but it is changed into sharp magnetic Bragg peaks below T_N. This indicates that strong spin fluctuation arising from geometrical frustration exists in Mn_3O_4 above T_N, which scatters acoustic phonons and is responsible for the increase in the thermal resistivity above T_N, whereas the spin fluctuation is suppressed below T_N, resulting in the decrease in the thermal resistivity.

Figure 2(b) shows the magnetic-field (H) dependence of the thermal resistivity ρ for FeV_2O_4. The magnetic field is applied parallel to the direction of the thermal current for all the measurements below. Unlike MnV_2O_4, the H dependence of ρ is barely observed at around $T_1 = 140$ K, $T_2 = 105$ K, $T_3 = 70$ K, or at lower T. As shown in Fig. 2(a), ρ sharply decreases with decreasing T below T_3, but the change in the magnetization M at T_3 is relatively small. This is in contrast to MnV_2O_4, for which ρ sharply decreases (because of the orbital ordering) and M sharply increases (because of the ferrimagnetic ordering) at $T_c = 57$ K, and thus, ρ sharply decreases with increasing H at around T_c [37].

On the other hand, the thermal resistivity ρ of CoV_2O_4 increases with increasing H at low T, as shown in Fig. 3(b). The change in the thermal resistivity ($\Delta\rho$) with an applied magnetic field of 5 T is plotted as a function of T in Fig. 3(c). $\Delta\rho$ increases with decreasing T below 40 K, though an anomaly is barely observed at around $T_N = 170$ K. This H dependence of the thermal resistivity is similar to that of Al-doped MnV_2O_4, in which there is only a ferrimagnetic ordering at $T_N = 57$ K. It should be pointed out again that a divergent increase in ρ itself with decreasing T is similarly observed in these two compounds. It was previously shown [11] that the orbital fluctuation of the V t_{2g} states is enhanced as T decreases and approaches the orbital ordering temperature, and if the orbital ordering is coupled with the magnetism, the ordering temperature can be increased and the orbital fluctuation above the transition temperature can be enhanced by increasing the magnetic field. In CoV_2O_4 and Al-doped MnV_2O_4, the hypothetical orbital-ordering temperature is less than 0 K, but it can be increased by increasing the applied magnetic field. This can explain the increase in the thermal resistivity with the applied magnetic field at low T.

Mn_3O_4 exhibits a peculiar H dependence of the thermal resistivity ρ. As shown in Fig. 5, the H dependence of ρ at various temperatures is not monotonic and has distinct anomalies, as shown by symbols. For example, at 39 K, ρ increases with $|H|$ for $|H| < 0.5$ T but decreases for

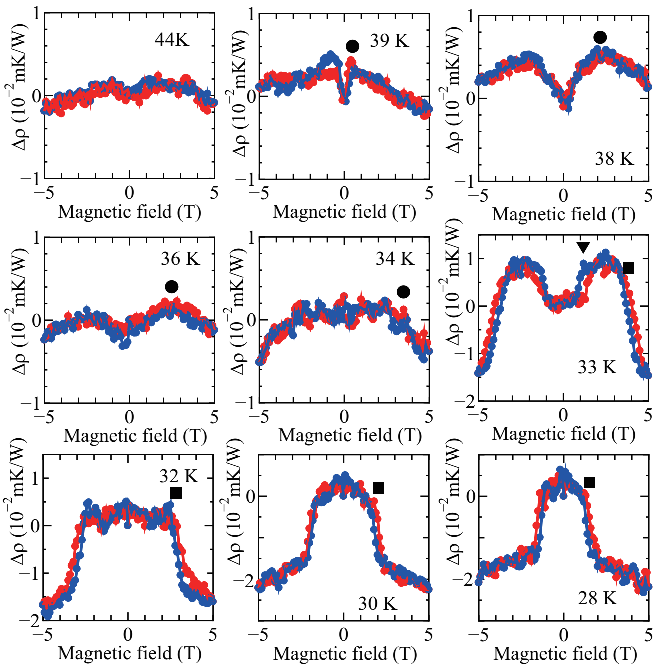


FIG. 5. (Color online) Magnetic-field dependence of the thermal resistivity at various temperatures for Mn_3O_4.

$|H| > 0.5$ T with a clear anomaly at 0.5 T shown by a closed circle. The position of this anomaly shifts to a higher H with decreasing T. We plot the positions of the anomalies in the H-T space in Fig. 4(b). We found that these anomalies lie on the phase boundaries of Mn_3O_4 when H is applied along the [100] direction reported by Nii *et al.* [32]. Namely, the circles correspond to the boundary between the Yafet-Kittel phase and the incommensurate phase, the triangle corresponds to that between the incommensurate phase and the cell-doubled phase, and the squares correspond to that between the cell-doubled phase and the Yafet-Kittel orthorhombic phase. The Yafet-Kittel orthorhombic phase, which appears only when a finite magnetic field is applied along the [100] direction, is similar to the Yafet-Kittel phase between T_N and T_1, except for the fact that it is orthorhombic (the in-plane axis along the H direction is longer than that along the other directions) and its total magnetic moment is along the [100] direction [32]. These results indicate that the change in the magnetic configuration affects the thermal resistivity in Mn_3O_4. Note that the reduction of ρ from the cell-doubled phase to the Yafet-Kittel orthorhombic phase (squares in Fig. 5), which amounts to several tens of percent, is the largest among the changes in ρ at various phase transitions induced by a magnetic field.

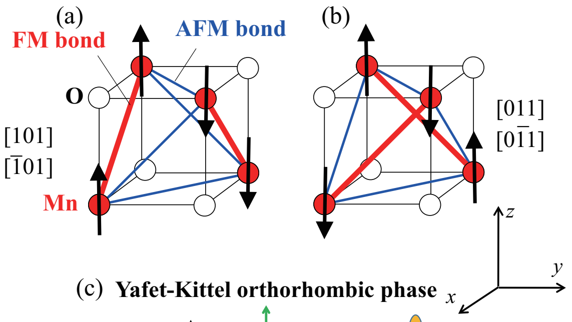
IV. DISCUSSION

The absolute values of the thermal resistivity ρ in the paramagnetic phase of spinel vanadates, 0.3–0.4 mK/W, are comparable to those of perovskite vanadates ($R\mathrm{VO}_3$) [4] and titanates ($R\mathrm{TiO}_3$) [5], in which two electrons and one electron, respectively, exist in the triply degenerate t_{2g} states, and thus an orbital degree of freedom exists in the transition metals similarly to in spinel vanadates. On the other hand, the absolute values of ρ in the paramagnetic phase of Mn_3O_4, ~0.1 mK/W, are similar to those of perovskite manganites ($LaMnO_3$) [3], where the degeneracy of the doubly degenerate e_g states in Mn^{3+} ions is lifted at several hundred K. Note that the absolute values of ρ in hexagonal $YMnO_3$, in which the Mn^{3+} ion is surrounded by five oxygen ions and the $3d^4$ electrons in the Mn^{3+} ion take a nondegenerate ground state, are also ~0.1 mK/W [2]. These results suggest that the absolute values of thermal resistivity (conductivity) can be a good measure of the strength of orbital fluctuation when compounds with the same crystal structure are discussed and compared.

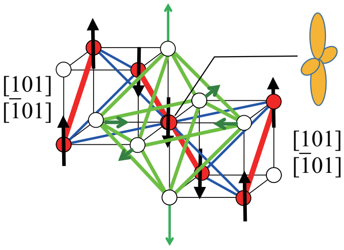
Next, let us discuss the behaviors in the ordered phase. The results for spinel vanadates indicate that orbital ordering suppresses the thermal resistivity, in contrast to antiferromagnetic ordering. For the cubic spinel AB_2O_4, in which both the A and B sites are occupied by magnetic ions, it is known that various types of ferrimagnetic ordering undergo energetic competition and the component of the magnetic moment perpendicular to the direction of the ferrimagnetic moment tends to fluctuate because of geometrical frustration [38]. Such spin fluctuation can be suppressed upon orbital ordering, which introduces anisotropy into the magnetic interactions and suppresses the geometrical frustration. The change in the magnetic structure at T_3 in FeV_2O_4 can be interpreted in this context. We speculate that the remnant spin fluctuation in the ferrimagnetic phase without orbital ordering is the reason for the thermal resistivity being hardly suppressed at a purely magnetic phase transition in spinel vanadates.

On the other hand, the magnetic ordering substantially affects the thermal resistivity of Mn_3O_4, which has a tetragonal distortion. Nii *et al.* pointed out [32] that in this compound, both the Yafet-Kittel phase and the cell-doubled phase are characterized by a local tetrahedron whose four corners are occupied by two up-spin Mn^{3+} ions and two down-spin Mn^{3+} ions, which are canted toward the [110] direction because of the antiferromagnetic coupling with Mn^{2+} ions in the A site (whose spins are along the [110] direction). Among the six Mn^{3+}-Mn^{3+} bonds in such a tetrahedron, four are antiferromagnetic bonds and two are “wrong” ferromagnetic bonds in terms of the antiferromagnetic interaction between Mn^{3+} spins. Since the ab axis is shorter than the c axis in the tetragonal phase, the shorter bonds along the ab plane ([110] and [$1\bar{1}0$] directions) should be antiferromagnetic bonds. There are still two possible configurations of the up spins and down spins in the tetrahedron; as shown in Figs. 6(a) and 6(b), ferromagnetic bonds can be along the [101] and [$10\bar{1}$] directions or along the [011] and [$01\bar{1}$] directions. Such degeneracy of the spin configurations is responsible for the relatively low T_N in Mn_3O_4. The Yafet-Kittel phase and cell-doubled phase can be regarded as two different arrangements of these tetrahedra. It is reasonable that the spin fluctuation existing above T_N, which arises from the two energetically degenerate spin configurations in each tetrahedron, is gradually suppressed during the successive magnetic transitions from the Yafet-Kittel, incommensurate, and cell-doubled phases, resulting in a decrease in the thermal resistivity.

Furthermore, it has recently been shown that the successive magnetic phase transitions and those with applied magnetic field in Mn_3O_4 are accompanied by structural distortions



(c) **Yafet-Kittel orthorhombic phase**



(d) **Cell-doubled phase**

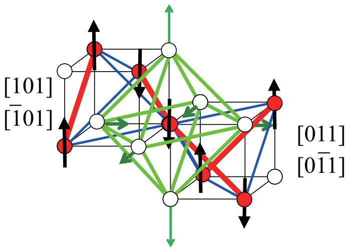


FIG. 6. (Color online) (a), (b) Two possible configurations of the Mn^{3+} spins on the corners of a tetrahedron in Mn_3O_4. Note that the canting of the spins toward the [110] direction is ignored. (c), (d) Spin configurations (black arrows) and resultant distortions of MnO_6 octahedra (green arrows) in (c) the Yafet-Kittel orthorhombic phase and (d) the cell-doubled phase. For all the figures, thicker red lines correspond to longer ferromagnetic bonds and thinner blue lines correspond to shorter antiferromagnetic bonds.

[30–34], suggesting that the orbital degrees of freedom of the e_g states in the Mn^{3+} ions play some roles in this compound. Nii *et al.* pointed out [32] that the exchange striction, by which the antiferromagnetic Mn^{3+}-Mn^{3+} bonds become shorter than the ferromagnetic bonds, induces the distortion in the MnO_6 octahedra in the magnetically ordered phase and causes the hybridization of the unoccupied $x^2 - y^2$ state with the occupied $3z^2 - r^2$ state for the Mn^{3+} e_g states. In this process, the sign of the hybridized state depends on the direction of the distortion in the MnO_6 octahedra along the *ab* plane (elongation or contraction). However, in the cell-doubled phase, the distortion of some of the MnO_6 octahedra is not a simple elongation or contraction, as illustrated in Fig. 6(d), and thus it cannot determine the sign of the hybridized state. In the orthorhombic Yafet-Kittel phase, however, the distortion of all the MnO_6 octahedra is simple elongation/contraction [Fig. 6(c)], and thus it can determine the sign of the hybridized states. [The orbital shape of the occupied *e* state is illustrated in Fig. 6(c).] The large suppression of the thermal resistivity from the cell-doubled phase to the orthorhombic Yafet-Kittel phase with an applied magnetic field can be explained by such a suppression in the orbital fluctuation of the hybridized e_g states.

V. SUMMARY

We studied the thermal resistivity (inverse thermal conductivity) of three transition-metal oxides with the spinel structure, FeV_2O_4, CoV_2O_4, and Mn_3O_4, and compared the results with those of previously reported for MnV_2O_4. We found that for the compounds having V^{3+} ions, the thermal resistivity decreases with the orbital ordering of V^{3+} ions, whereas it does not exhibit any anomaly upon purely magnetic ordering. This indicates that the orbital fluctuation of the triply degenerate t_{2g} states occupied by two electrons in V^{3+} ions dominates the thermal resistivity of these compounds. On the other hand, for Mn_3O_4, in which the orbital degeneracy of the e_g states in Mn^{3+} ions is lifted at high temperatures because of the Jahn-Teller distortion, the thermal resistivity increases with decreasing T above T_N but decreases with successive magnetic phase transitions. This can be interpreted in such a way that Mn^{3+} spins having antiferromagnetic interactions with each other on the corner of a tetragonally distorted tetrahedron give rise to two possible energetically degenerate spin configurations, resulting in the increase in the spin fluctuation above T_N and its decrease with successive magnetic phase transitions. We also found that the Yafet-Kittel orthorhombic phase, which appears when a magnetic field is applied along the [110] direction (in the cubic setting), has the lowest thermal resistivity among the various phases. This is caused by the ordering of the $x^2 - y^2$ state hybridized with the $3z^2 - r^2$ state with a specific sign in this phase.

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