4C, 16A2/2005S2-003

Coupling between magnetism and crystal structure in spinel MnV_2O_4

Takehito SUZUKI[1], Michitaka KATSUMURA[1], Kouji TANIGUCHI[2], Taka-hisa ARIMA[2], Takuro KATSUFUJI[1,*]

[1]Dep. Phys., Waseda University, Tokyo 169-8555, Japan
[2]IMRAM, Tohoku University, Sendai 980-8577, Japan

Introduction

Orbital degrees of freedom arise when degenerate states ($3d$ orbitals, for example) with the number of degeneracy n are occupied by a number of electrons other than n or $2n$. This orbital degree of freedom often dominates the crystal structure and the elastic properties of materials. For example, orbital-ordered states, in which particular orbitals are occupied by electrons at each site, lead to lattice distortion in lower crystal symmetries because of the anisotropy of d orbitals. In addition, the orbital degree of freedom is often coupled with the spin degree of freedom. This coupling arises from the fact that the occupation of specific orbitals favors specific types of magnetic interaction (the so-called Kugel-Khomskii type coupling). Such oribital-spin coupling gives rise to intriguing magnetic-field effects in transition-metal oxides.

We studied spinel MnV_2O_4 as a prototypical system having the orbital degree of freedom in a cubic structure. In this compound, the A site of the spinel structure (AB_2C_4) is occupied by the Mn^{2+} ion, which is in the $3d^5$ high-spin configuration with no orbital degree of freedom, and can be regarded as a simple $S=5/2$ spin. On the other hand, the B site is occupied by the V^{3+} ion, which takes the $3d^2$ high-spin configuration in the triply degenerate t_{2g} orbital, and has the orbital degree of freedom. It is known that this compound exhibits a structural phase transition from cubic to tetragonal at 57 K, and also ferrimagnetic ordering at the same temperature [1]. We performed the x-ray diffraction measurement of single crystals of MnV_2O_4 to clarify the orbital state of this compound [2].

Results

Figure 1 (a) shows the temperature dependence of the (440) peak (in the cubic setting) of MnV_2O_4. The peak sharply splits into two peaks below 57 K, indicating the structural phase transition from cubic to tetragonal at this temperature. The difference of the shorter and longer axes amounts to more than 1%. Figure 2 (b) shows the magnetic field dependence of the (800) peak (in the cubic setting) at 59 K, slightly above the transition temperature. Without magnetic field, the peak corresponds to that of the cubic phase. However, with applied magnetic field above 4 T, the peak jumps to that of the tetragonal phase, indicating the magnetic-field-induced structural phase transition from cubic to tetragonal. Anomalies of magnetization and strain were also observed at the same temperature and magnetic field. This experimental result demonstrates the coupling between magnetism and crystal structure in MnV_2O_4.
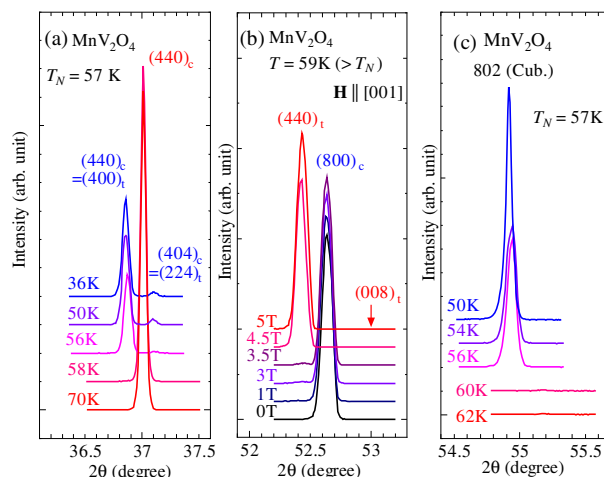

Fig. 1 (a) Temperature dependence of the (440) peak, (b) magnetic-field dependence of the (800) peak at 59 K, and (c) temperature dependence of the (802) peak in the x-ray diffraction.

Figure 1 (c) shows the (802) peak (in the cubic setting) of MnV_2O_4. This peak is forbidden in the cubic spinel structure by the diamond glide symmetry. However, this peak appears below 57 K, indicating that the diamond glides no more exist in the tetragonal phase of MnV_2O_4. This indicates the antiferro-type ordering of V t_{2g} orbitals, as proposed by Tsunetsugu and Motome [3], where the yz and the zx orbitals are alternately occupied along the c axis.

From these experimental results, we can conclude that there is orbital ordering in MnV_2O_4, which is strongly coupled with spin ordering. The mechanism of this simultaneous orbital and spin ordering can be explained by Kugel-Khomskii type coupling, in which the ferromagnetic alignment of V spins (which is the case in the ferrimagnetic state) favors the antiferro-type ordering of V t_{2g} orbitals [2].

References

[1] K. Adachi *et al.*, Phys. Rev. Lett. **95**, 197202 (2005)
[2] T. Suzuki *et al.*, Phys. Rev. Lett. **98**, 127203 (2007).
[3] H. Tsunetsugu and Y. Motome , Phys. Rev. B, **68**, 060405 (2003).

* katsuf@waseda.jp