Structural phase transitions and magnetic field effect on crystallographic domains in spinel oxide FeV₂O₄

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1 Introduction

Vanadium spinel oxide FeV₂O₄ is a unique compound because both of Fe²⁺ and V³⁺ ions have orbital degrees of freedom. This compound shows successive structural transitions (cubic \rightarrow tetragonal (c < a) \rightarrow orthorhombic \rightarrow tetragonal (c > a)) for polycrystalline sample with decreasing temperature [1]. Another transition from tetragonal (c > a) to orthorhombic at 35 K was reported [1], however, this transition was not observed by the other reports [2, 3]. These transitions have been discussed in terms of the orbital states, however, two orbital ordering (OO) models of the V^{3+} ions below 60 K were proposed. One is ferro-OO with the space group of $I4_1/amd$ by x-ray diffraction experiment [2] and another is antiferro-OO with the space group $I4_1/a$ by x-ray magnetic circular dichroism [4]. On the other hand, we have found the anomalous magnetic properties, i.e., the magnetization jumps in the temperature dependence of magnetization, and difference between the isothermal magnetization (M-H) curves for the zero-field cooled (ZFC) and field cooled (FC) conditions in the single crystal [5]. This may be caused by the difference in the crystallographic domain structure under the magnetic field, however, its origin has been unclear. In this study, we have carried out single crystalline x-ray diffraction experiments of FeV₂O₄ to investigate the crystal system and extinction rule of the Bragg reflections at the lowest temperature and the crystallographic domain structure under the magnetic field.

2 Experiment

Single crystals of FeV_2O_4 were grown by the floatingzone method in Ar gas flow. X-ray diffraction experiments were carried out using a four-circle diffractometer at BL-4C under zero magnetic field and a two-circle one at BL-3A under the magnetic field. In the measurement, the crystal was mounted so that a magnetic field is parallel to the [001] direction of the cubic axis and perpendicular to the scattering vector.

3 Results and Discussion

Figure 1 shows the temperature dependence of 800 Bragg reflection of the cubic spinel structure (θ -2 θ scan). Successive structural transitions from cubic to tetragonal (c < a) at ~140 K, from tetragonal to orthorhombic at ~100 K, and from orthorhombic to tetragonal (c > a) at ~60 K were observed. However, the structural transition from tetragonal (c > a) to another orthorhombic below 35 K was not observed even at 10 K as shown in Fig. 1.



Fig. 1: Temperature dependence of 800 cubic reflection.

We also examined appearance of the forbidden reflections of the space group $I4_1/amd$ such as 420 reflections below 60 K, which are observed in the case of the space group $I4_1/a$. However, the forbidden reflections could not be detected. This result indicates that the OO model of the V³⁺ ions is the ferro-OO.

Figure 2(a) shows the diffraction profiles of 800 cubic Bragg reflection at 5 K and 50 K under the magnetic field of 0.5 T. Before measurement, the sample was cooled down to 5 K without a magnetic field and followed by applying the magnetic field. Two peaks corresponding to 008 and 800 reflections were observed at 5 K regardless of application of the magnetic field of 0.5 T, however, 008 reflection disappears in warming up to 50 K. This result indicates that the magnetic domains correspond to the crystallographic ones and that the multi-domain state changes into single-domain one under the magnetic field by warming. This fact is consistent with the observed in the temperature magnetization jump dependence of magnetization [5]. Figure 2(b) shows the

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Fig. 2: (a) 800 cubic Bragg reflection under the magnetic field of 0.5 T at 5 K and 50 K. (b) Magnetic field dependence of 800 cubic Bragg reflection at 5 K corresponding to ZFC condition.

magnetic field dependence of 800 cubic Bragg reflection at 5 K. After cooling down to 5 K without the magnetic field, the measurement was carried out with increasing the magnetic field up to 5 T followed by decreasing it down to 0 T, which is corresponding to the ZFC condition. 008 reflection was observed even under the magnetic field of 1 T, indicating the multi-domain state. Under the high magnetic field of 5 T, the crystal forms the singledomain state, therefore, high magnetic field may forcibly induce the single-domain state. When the magnetic field decreases down to 0 T, 008 peak appears, which leads to that the magnetic domain structure is reconstructed from the single-domain to multi-domain state in order to decrease the magnetostatic energy. These results are consistent with the fact that the M-H curve in the ZFC condition has a large coercive field [5].

References

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