

# Critical Phenomena in Helical Magnet $\beta$ -MnO<sub>2</sub>: X-ray Magnetic Scattering Study

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## Abstract

It has been theoretically expected that critical phenomena of helimagnetism is drastically different from those of collinear antiferromagnetism due to the existence of *chiral* degree of freedom. In order to verify this prediction, we carried out X-ray magnetic scattering measurements on a simplest helimagnet  $\beta$ -MnO<sub>2</sub> using a synchrotron radiation. The critical exponent  $\beta$  was estimated at  $0.25 \pm 0.05$  from the temperature dependence of the scattering intensity. This value is consistent with the predicted value 0.253 for chiral system rather than with 0.346 for collinear system.

*Key words:* critical phenomena; manganese oxides; helical magnetism; X-ray magnetic scattering

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## 1. Introduction

Critical phenomenon is one of the most important concepts in the physics of phase transition. In the vicinity of the critical temperature, it is known that various physical quantities exhibit singular behaviors. In antiferromagnets, the local magnetization  $M$  follows the function  $M = A(T_N - T)^\beta$ , where  $A$  and  $T_N$  are a constant and the Neel temperature, respectively. The scaling theory predicts that the critical exponent  $\beta$  should be a universal value dependent only on the *universality class* of the system and independent of the other detail. Universality class depends on the dimensionality of the system, spin anisotropy, *etc.* Kawamura has pointed out [1–3] that also *chirality* makes definite difference to the universality class. A helimagnet has chiral degeneracy because a clockwise helix and an anticlockwise helix are equally desirable. Therefore, the

critical exponents in a helimagnet are expected to be very different from those in collinear antiferromagnets.

The title compound  $\beta$ -MnO<sub>2</sub> has been known as a simplest helimagnet,[4] where there are only short-range exchange interactions. However, there have been no studies on its critical phenomenon because of the difficulty in the growth of single crystals large enough for neutron diffraction experiments. For a small crystal, X-ray magnetic scattering[5,6] sometimes has the advantage of neutron magnetic scattering if a very strong X-ray source such as a synchrotron radiation is available. We have already observed X-ray magnetic scattering in  $\beta$ -MnO<sub>2</sub> using a synchrotron radiation generated by a bending magnet in KEK, Tsukuba, Japan.[7] However, the scattering intensity was insufficient for the estimation of the critical exponent  $\beta$ . This time, we used a very strong synchrotron radiation generated by an undulator in SPring-8, Hyogo, Japan. This is the first report on the critical phenomenon of  $\beta$ -MnO<sub>2</sub>.

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## 2. Result and Discussion

Single crystals of  $\beta$ -MnO<sub>2</sub> were obtained using a hydrothermal method. A crystal ( $3 \times 0.6 \times 0.6$  mm<sup>3</sup> size) is fixed in a cryostat mounted on a four circle diffractometer. The temperature was stabilized by using a PID controller. The drift of the temperature during the data collection was  $\pm 10$  mK. A monochromized X-ray beam generated at the experimental station BL46XU of SPring-8 was used for the measurement. The energy of the X-ray was 22.154 keV.

Below  $T_N$  ( $\approx 94$  K), several magnetic scattering satellites appeared. For the estimation of the critical exponent, we collected the data of the (1 0 2- $\eta$ ) satellite, where  $\eta \approx 0.3$ . First we fitted the scattering intensity with the function of the temperature  $a(b-T)^c$ , where  $a, b$  and  $c$  are fitting parameters. The optimized values of  $b$  and  $c$  are 94.73 and 0.421, respectively. The value  $b = 94.73$  corresponds to  $T_N$  measured by the present thermometer. Because the scattering intensity is proportional to the square of the local magnetization  $M$ , we can roughly estimate  $\beta$  at  $c/2 = 0.21$ . For more exact estimation of  $\beta$ , we show a log-log plot of the scattering intensity versus  $(T_N - T)/T_N$  in Fig. 1, where we use 94.73 K as  $T_N$ . From the slope of the figure, we can estimate  $\beta$  at 0.26. Because of the poor S/N ratio, it is difficult to give more precise value of  $\beta$  at present. However, we can conclude that the value of  $\beta$  lies between 0.2 and 0.3.

We compare the obtained value of  $\beta$  with the theoretical prediction. It is known that the network of exchange interaction is three-dimensional and that the spins have XY anisotropy in  $\beta$ -MnO<sub>2</sub>[8,9]. First, let us try the XY collinear model, ignoring the chirality. In this case the universality class is  $V = S_1 = \text{SO}(2)$  and the predicted value of  $\beta$  is 0.346, significantly larger than our observed value. Then, let us take the chirality into account. In this case the universality class becomes  $V = Z_2 \times S_1 = \text{O}(2)$ . The Monte Carlo simulation by Kawamura[2] shows the  $\beta$  is  $0.253 \pm 0.01$ . This agrees with our observation.

The critical exponents have already been examined in rare-earth metal helimagnets such as Tb, Dy and Ho. However, their experimental values of  $\beta$  scatter from 0.21 to 0.39, depending on the method and on the research group. Kawamura has attributed[3] this to the long-range character of the RKKY-type exchange interaction which is responsible for the helimagnetism of rare-earth metals. Generally, the range of RKKY interaction is of the order of the wavelength of the helix. If the magnetic correlation length becomes shorter than the range of RKKY interaction, a mean-field like behavior ( $\beta = 0.5$ ) appears and the effect of chirality disappears. Therefore,  $Z_2 \times S_1$  ( $\beta = 0.253$ ) behavior is limited in a very narrow temperature region around  $T_N$ .

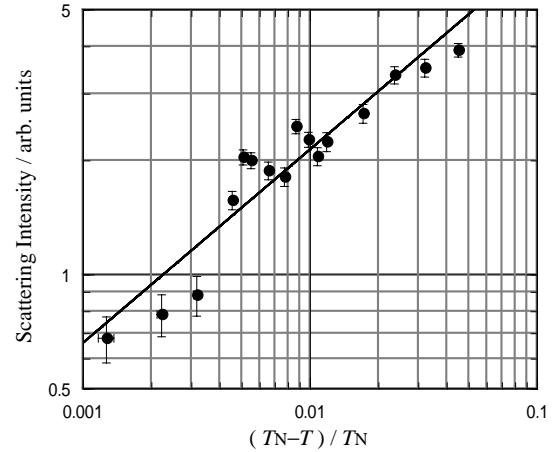


Fig. 1. The log-log plot of the scattering intensity versus normalized temperature.

This makes it difficult to determine the critical exponent of the rare-earth helimagnets. On the other hand,  $\beta$ -MnO<sub>2</sub> is an ideal helimagnet for the study of critical phenomena because the helimagnetism is caused by only short-range exchange interactions.[4]. Therefore, we can use wider temperature region for the estimation of  $\beta$ .

## 3. Conclusion

We measured the critical exponent  $\beta$  of an ideal helimagnet  $\beta$ -MnO<sub>2</sub> using X-ray magnetic scattering for the first time. The obtained value  $0.25 \pm 0.05$  support the  $Z_2 \times S_1$  universality-class model. This indicate that the chiral degeneracy has a decisive effect on the critical phenomenon.

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