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Magnetic Neutron Scattering of the Frustrated System $ZnCr_2O_4$

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$ZnCr_2O_4$ has a typical normal spinel structure with magnetic Cr^{3+} ions only at the B-sites. The magnetic moments of these ions frustrate each other by the nearest neighbor antiferromagnetic (AF) interaction. This frustration makes the magnetic structure of this oxide complex. In order to investigate the magnetic structure of $ZnCr_2O_4$, we carried out a neutron diffraction study with a single crystal. Magnetic diffuse scattering spread widely in reciprocal space was observed. Its intensity gradually increased with the decrease of temperature below 100 K and abruptly dropped aroud 10 K. Accompanying this drop, AF Bragg reflections at (h/2,k,l) and (h/2,k/2,l) appeared rapidly at 10 K($= T_N$) indicating a first order transition. However, successive magnetic transitions observed in the similar normal spinel $MgCr_2O_4$ do not occur in $ZnCr_2O_4$. These results suggest that the AF long range order emerges below T_N due to the reduction of the spin frustration. This reduction seems to be associated with the structural phase transition indicated by dielectric measurements.

KEYWORDS: $ZnCr_2O_4$, frustrated system, spinel

§1. Introduction

The magnetic transitions of frustrated systems are interesting because of their variety. In magnetic spinel compounds, the collinear Néel state is generally realized because the A - B antiferromagnetic interaction is stronger than the B - B antiferromagnetic interaction. In a series of spinels with Zn ions at the A - site, the weak A - B antiferromagnetic interaction makes its spin configuration complex. $ZnCr_2O_4$ is a typical substance of such spinels with magnetic ions (Cr^{3+}) only at the B-sites.[1)] The network of the B-site in the spinel structure is composed of tetrahedra sharing their corners with four other neighboring tetrahedra. Consequently, magnetic ions are frustrated with respect to the antiferromagnetic interaction between the nearest neighbors.[2)] In the case of the isomorphous spinel $MgCr_2O_4$, two distinct transitions to magnetically ordered states were observed at 16 K and 13.5 K. Magnetic diffuse scattering was observed at 20 K and it indicates the existence of frustration in $MgCr_2O_4$.[3)] The reflections associated with the 16 K transition were explained by a noncollinear antiferromagnetic structure with the magnetic unit cell identical with the chemical cubic unit cell. The low temperature reflections below the 13.5 K transitions were explained by either of two nonequivalent noncollinear antiferromagnetic structures.

In the susceptibility measurement of $ZnCr_2O_4$ the Curie-Weiss type temperature dependence was reported only higher temperature region above 150 K , much higher than its antiferromagnetic transition temperature T_N (=10K).[4)] Its asymptotic Curie temperature, θ is estimated to be - 330 K and its Curie constant leads to a spin of S=3/2 , corresponding to a localized Cr^{3+} ion. This deviation of susceptibility from the Curie-Weiss behavior below 150 K might be caused by the frustration. A softening of elastic constants in a sound velocity measurement was reported and it indicates a crystallographic transition at T_N.[4)] Recently, S.-H. Lee et. al. claimed that this transition is a three dimensional analog of the spin - Peierls transition from the inelastic magnetic neutron scattering data.[5)] In this connection, it is interesting to know what magnetic structure is realized in $ZnCr_2O_4$ and how it transforms into an ordered state.

§2. Experiment and Result

The single crystal sample of $ZnCr_2O_4$ was grown by a flux method, in a form of octahedron about 3 mm on edge. The neutron scattering data were collected in a temperature range between 2 K and 15 K with the PONTA (Polarized Neutron Triple-Axis Spectrometer) installed at JRR-3M in JAERI. The single crystal sample was fixed as the [001] or [011] direction of high temperature cubic phase to be vertical to the horizontal plane.

In a few experimental runs the sample was field cooled from about 30 K to 2 K in a magnetic field of 50 kOe applied along the [100] or [010] direction. If this magnetic field cooling procedures makes a change of its domain distribution it is expected that the difference of the intensity of a pair of magnetic peaks such as (1, 3/2, 0) and (3/2, 1, 0) .

Figure 1 shows the positions of the magnetic Bragg reflection observed at 2 K and the regions observed diffuse scattering above T_N. Magnetic Bragg peaks with both (h/2, k, l) and (h/2, k/2, l) indices were observed. The

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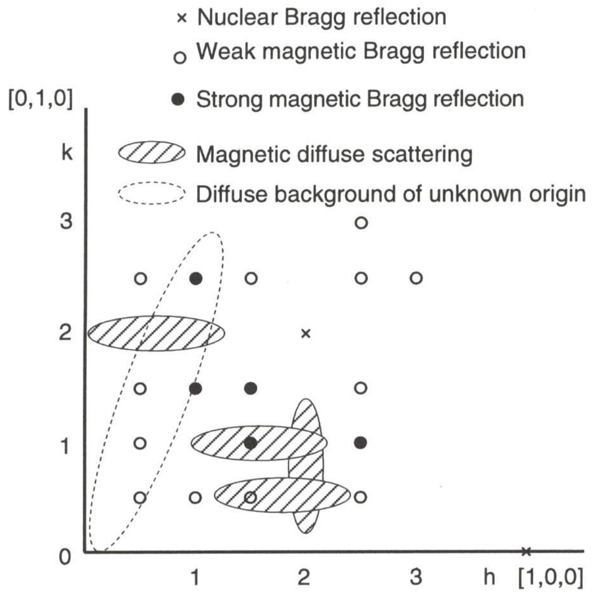

Fig. 1. Positions of magnetic Bragg reflections and magnetic diffuse scattering.

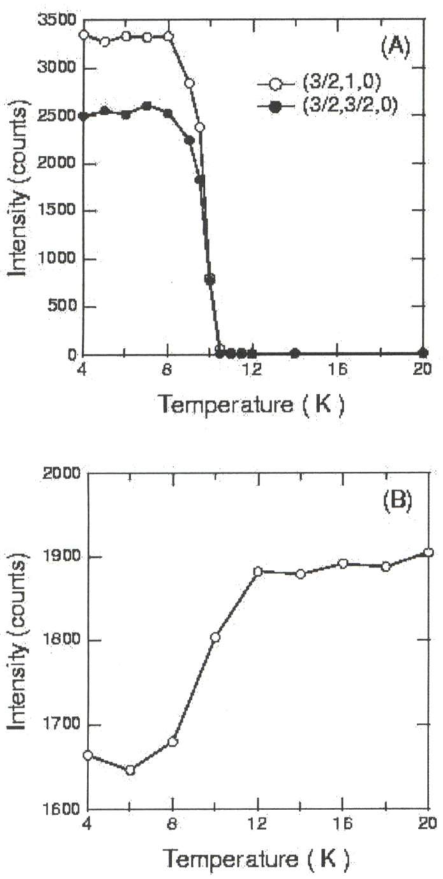

Fig. 2. Temperature dependence of the magnetic Bragg peak intensities at (3/2, 1, 0) and (3/2, 3/2, 0) (A), and that of magentic diffuse scattering around (2,1,0) (B)

temperature dependences of (3/2, 1, 0) and (3/2, 3/2, 0) reflections are shown in Fig. 2(A). They had a similar temperature dependence and vanished at 10 K, which is identified as the Néel temperature T_N. Thus, two successive magnetic transitions claimed on $MgCr_2O_4$[3] is denied in the case of $ZnCr_2O_4$. Magnetic diffuse scattering was observed at the hatched regions in Fig.1. Its intensity gradually increased with decreasing temperature from 100 K down to T_N. Figure 2(B) shows the temperature dependence of diffuse scattering intensity studied around the reciprocal lattice point (2, 1, 0) around T_N. These results indicate the presence of a remarkable magnetic short range order in much higher temperature than T_N. A deviation in the susceptibility data from the Curie-Weiss law and a low-frequency dielectric dispersion were reported in the same temperature range. The diffuse scattering intensity shows a rapid drop around T_N, and remaines at a finite level below T_N. The intensity of the diffuse scattering widely distributes in the reciprocal space and this distribution seems to be independent of temperature.

To decide the magnetic structure of the ordered state, it is indispensable to use a single crystal sample composed of a single antiferromagnetic domain. However, the magnetic field cooling did not make any influence in the diffraction pattern within the accuracy of the experiment. If the ordered phase has a certain magnetic symmetry, it is possible that a single domain state can be obtained by cooling a single crystal sample with both magnetic and electric fields applied simultaneously.

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