Magnetic structure of Ba$_2$Mg$_2$Fe$_{12}$O$_{22}$ under the magnetic field

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Abstract. Ba$_2$Mg$_2$Fe$_{12}$O$_{22}$ has a strong coupling between electric polarization $P$ and magnetization $M$. To understand the multiferroic properties of the system, we performed crystal structural and precise magnetic structural analyses of Ba$_2$Mg$_2$Fe$_{12}$O$_{22}$ under a magnetic field ($T = 4K$, $H = 0.3T$) using 4 circle neutron diffractometer (FONDER) at JRR-3M Tokai. We collected 105 points of magnetic scattering for $q = (0,0,3/2)$ and over 150 points of fundamental scattering which contains ferri magnetic component. Using the part of these data, we reinvestigate magnetic structure of Ba$_2$Mg$_2$Fe$_{12}$O$_{22}$, in which 14 (Mg,Fe) sites have been included.

1. Introduction

Recently, it has been established that the ferroelectricity is coupled with the spiral magnetic structure under an applied magnetic field.

The ferroelectricity induced by magnetic order mostly in frustrated magnets are nowadays referred to as magneto-electric (ME) multiferroic, or often only as multiferroic. Some distinct types of microscopic origins relevant to the spin-driven ferroelectricity are discussed in detail.

The typical multiferroic materials, characterized by a helimagnetic order giving rise to ferroelectricity, has been of great interest, after the discovery of a giant magnetoelectric effect in TbMnO$_3$ [1].

Kimura et al.[3] discovered ferroelectric polarization in a fan phase with a magnetic modulation wave number $q = (0 \ 0 \ 3/2)$ in Ba$_2$Mg$_2$Fe$_{12}$O$_{22}$.
Figure 1. (a) and (b) are schematic illustrations of spin structures proposed by Ishiwata *et al.* for ferroelectric phase. (c) is ferromagnetic component of these model. (d) and (e) are $k=(0,0,\frac{3}{2})$ magnetic component of (a),(b) model respectively.

A similar material Ba$_{2-x}$Sr$_x$Zn$_2$Fe$_{12}$O$_{22}$. However, since the proposed fan structure does not break the space inversion, the mechanism which induces ferroelectric polarization is not yet solved.

The magnetic structure of Ba$_{2-x}$Sr$_x$Zn$_2$Fe$_{12}$O$_{22}$ has been studied using neutron diffraction [4,5,6]. Momozawa *et al.* assumed the two blocks L and S as units of the magnetic structure of this system. A block L possesses a large effective magnetic moment $\mu L$, and a block S possesses a small effective magnetic moment $\mu S$.

Ishiwata *et al.* proposed a new magnetic structure whereby a longitudinal conical structure undergoes a transition to a transverse conical structure which has the rotation axis perpendicular to the propagation direction as opposed to the longitudinal conical spiral, resulting in ferroelectricity [7].

However, neutron-diffraction studies for magnetic structure in each 14 (Mg,Fe) magnetic sites have not yet been conducted, therefore the precise source of ferroelectric polarization is not known. Hence, the determination of the magnetic structure in the Ba$_2$Mg$_2$Fe$_{12}$O$_{22}$ compound provides us important information for discussing a mechanism of ferroelectricity in the system.

2. Experimental

The sample preparation procedure and the method of the electric polarization $P$ measurement have been reported previously[8]. Magnetization $M$ was measured with a commercial dc superconducting quantum interference magnetometer at the Center for Low Temperature Science, Tohoku University, Japan. The magnetic field dependence of magnetization ($M$) and electric polarization ($P$) at 4.3K parallel to the $b$-axis is reported by Taniguchi *et al.*[8]. In this measurement, Ba$_2$Mg$_2$Fe$_{12}$O$_{22}$ shows the strongest electric polarization ($\sim 100$ C/m$^2$) under a magnetic field of 0.3T. Single crystal structure and magnetic structure analyses in a neutron diffraction measurement under the magnetic field were carried out on a four-circle neutron diffractometer (FONDER) installed in the guidehall of JRR-3M at the Japan Atomic Research Institute, Japan. The sample was mounted with its $b$ axis perpendicular to the scattering plane in a closed-cycle $^4$He refrigerator. An external magnetic field was applied parallel to the $b$ axis with a split-type electromagnet.
3. The magnetic structure analysis

We determine magnetic structure of \( k = (0, 0, \frac{3}{2}) \) and ferri magnetic component separately. We collected \( k = (0, 0, \frac{3}{2}) \) for magnetic satellite scattering and fundermental Bragg scattering for ferri magnetic components.

In this phase, we also observed magnetic scattering \( k = (0, 0, \frac{3}{4}) \), however the intensities of \( k = (0, 0, \frac{3}{4}) \) are much weaker than those of \( k = (0, 0, \frac{3}{2}) \), so we assume that the magnetic structure \( k = (0, 0, \frac{3}{2}) \) are main contribution to the ferroelectricity in this phase.

3.1. The antiferromagnetic component in the magnetic structure

We collected 105 points of magnetic scattering and determined the magnetic structure of 14 sites of (Mg,Fe) sites on three axis \( 14 \times 3 = 42 \). (2 ~ 3 times of data points are needed for precise magnetic structure analysis.) We assumed that 14 (Mg,Fe) sites have no magnetic moment along the \( b \)-axis and determined the magnetic structure after many trials of model fitting. The reliability factors are \( R_p = 0.0726 \) and \( R_{wp} = 0.0574 \). We define here, \( R_p = \sum_i |F_{obs}(i) - F_{cal}(i)|/\sum_i F_{obs}(i), R_{wp} = \left[ \sum_i w(i)\{F_{obs}(i) - F_{cal}(i)\}^2 / \sum_i w(i)F_{obs}(i)^2 \right]^{1/2}, w(i) = 1/(dF_{obs}(i))^2 \). Here, \( i, F_{obs}, F_{cal}, w \) are data number, observed structure factor, calculated structure factor, weighted factor respectively.

![Figure 2](image)

Figure 2. The magnetic structure of \( k = (0, 0, \frac{3}{2}) \) components at \( T = 4K, H = 0.3T \). Green, deep blue and watery blue atoms are Ba, (Mg,Fe), and O atoms respectively.

3.2. The ferrimagnetic component in the magnetic structure

Next, we collected more than 60 points of fundermental reflections superposed to the magnetic and nuclear scattering. We determined the crystal structure using the reflection data \( 2\theta \geq 60^{\circ} \) where the magnetic scattering is negligibly small enough because the magnetic form factor \( f(sin\theta/\lambda) \) is very weak. We determined the structural parameters, the extinction parameter, Debye-Waller factor and scale factor. Using the reflection data \( 2\theta \leq 60^{\circ} \) and this structural parameter \( (I_{mag} = I_{obs} - I_{cal}) \), we obtained magnetic intensities of ferri magnetic components. We assumed that 14 (Mg,Fe) sites have same magnetic moment \( \mu_{Fe} = 1.975\mu_B \) and determined the magnetic structure after many trials of model fitting. The reliability factors are \( R_p = 0.0903 \) and \( R_{wp} = 0.0923 \).
4. Result and Discussion
Many trials of model fitting with different initial guesses verified that the obtained results are one of the best minima. Although there are several solutions with almost similar residuals, we discuss here our results and previous results. Compared with previous reported, we define \( S \) blocks and \( L \) blocks which propagate along the \( c \)-axis,

\[
\mu_{S_1} = (a*, b, c*) = (+1.815, -1.975, -0.101) \mu_B, \\
\mu_{L_1} = (a*, b, c*) = (-2.927, +1.975, -1.311) \mu_B, \\
\mu_{S_2} = (a*, b, c*) = (-1.815, -1.975, +0.101) \mu_B, \\
\mu_{L_2} = (a*, b, c*) = (+2.927, +1.970, +1.311) \mu_B
\]

These effective moments run on along the \( c \)-axis. It is difficult for us to discuss quantitatively so far, our results shows tranverse fan structure and longitudinal fan structure coexist which may produce ferroelectricity in \( \text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22} \).

Further research in this phase are now in progress. We will identify the best model to explain the experiments using a least-square method and the origin of spin driven ferroelectricity.

References
[1] Kimura T, Goto T, Shintani H, Ishizaka K, Arima T and Tokura Y, 2003 Nature 426 55
[2] Katsura H, Nagaosa N, and Balatsky A V 2005 Phys. Rev. Lett. 95 057205
[3] Kimura T, Lawes G and Ramirez A P 2005 Phys. Rev. Lett. 94 137201
[4] Momozawa N, Yamaguchi Y, Takei H and Mita M 1985 J. Phys. Soc. Jpn. 54 771
[5] Momozawa N, Yamaguchi Y, Takei H and Masaru Mita 1985 J. Phys. Soc. Jpn. 54 3895
[6] Momozawa N and Yamaguchi Y 1993 J. Phys. Soc. Jpn. 62 1292
[7] Ishiwata S, Okuyama D, Kakurai K, Nishi M, Taguchi Y and Tokura Y 2010 Phys. Rev. B 81 174418
[8] Taniguchi K, Abe N, Ohtani S, Umetsu H and Arima T 2008 Appl. Phys. Exp. 1 031301(1)-(3)