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Pyroelectric measurements on a geometrically frustrated spin system CuFeO$_2$ in pulsed high magnetic fields

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Abstract. Dielectric polarization $Pem[110]$ of a single crystal of a triangular lattice antiferromagnet CuFeO$_2$ has been measured by pyroelectric technique in pulsed high magnetic fields up to 43 T applied along the [001] direction. Among the five phases successively appeared in magnetic fields, a finite $P$ is observed only in the non-colinear incommensurate (5-sublattice like) phase. The present results indicate that no inversion symmetry breaking occurs in the 4th field-induced phase where a non-colinear spin structure is expected.

1. Introduction

Magnetoelectric effects are the phenomena in which the magnetization and the electric polarization are mutually correlated with each other in materials, and have been studied for several decades [1][2][3][4]. In general, inversion symmetry of the system must be broken to induce a spontaneous dielectric polarization. Recently, dielectric anomalies on geometrically frustrated spin systems have attracted much interest. Since various magnetic phases appear in a geometrically frustrated spin system in magnetic fields, some of those may lack the inversion symmetry, allowing the appearance of a spontaneous electric polarization. One possibility is a spin current mechanism due to spin-orbit interactions as recently predicted [5]. Here, a local polarization expressed as $p \propto -e_{12} \times (S_1 \times S_2)$ is predicted, where $e_{12}$ is the vector connecting the spin $S_1$ and the spin $S_2$ interacting with each other. In this situation, a finite dielectric polarization may appear, if the spins are arranged noncollinearly each other as is often the case in frustrated systems. Thus, a geometrically frustrated spin system is a good candidate to exhibit magnetoelectric effects.

Delafossite type quasi 2-dimensional triangular antiferromagnet CuFeO$_2$ is a typical geometrically frustrated spin system consisting of Fe$^{3+}$ ions [6]. Since the Fe$^{3+}(3d^{5})$ ion has no orbital degree of freedom, this compound is considered to be an ideal Heisenberg spin system. This compound is known to exhibit successive magnetic-field-induced phase transitions. When magnetic field is applied along the [001] direction at 1.6 K, five magnetic phases, i.e. 4-sublattice, 5-sublattice-like, 5-sublattice, 3-sublattice and 4th-field-induced (4th) phases appear successively [6][7]. The 4-, 5- and 3-sublattice phases are known to show commensurate and...
collinear spin structures, where the magnetization curve exhibits plateaus \[^8\][^9][^10]. On the other hand, incommensurate and noncollinear spin state is realized in the 5-sublattice-like phase \[^9\]. In this phase, the magnetization increases linearly with magnetic field \[^7\]. The 4th phase, which appears at high fields above 32 T, also exhibits a linear increase of the magnetization, suggesting a non-collinear spin configuration. The spin structure in the 4th phase is, however, still unknown.

Kimura \textit{et al.} have recently studied the magnetoelectric effect on CuFeO\(_2\) in dc magnetic fields up to 14 T, and found that spontaneous dielectric polarization appears in the non-collinear 5-sublattice-like phase but not in the collinear 4- and 5-sublattice phases \[^11\]. They concluded that the non-collinear spin structure plays a key role to induce electric polarization. Stimulated by their results, we have examined the magnetoelectric effect on CuFeO\(_2\) in very high magnetic fields, with attention being focused on the 4th phase where a non-collinear spin state is expected. To reach the 4th phase, pulsed high magnetic fields are needed. As far as we know, no magnetoelectric measurement has been done in pulsed fields. For this purpose, we have developed a dielectric polarization measurement technique in pulsed magnetic fields.

![Figure 1. The electrical circuit for the detection of the pyroelectric current.](image)

2. Experimental
A single crystalline sample was prepared by a floating zone method and cut into a thin plate (0.46×5×3 mm\(^3\)) with the largest surfaces perpendicular to the [110] direction. Silver paste (Dupon, 4922N) was used to make the electrodes as shown in Fig. 1. Strong magnetic fields up to 43 T were generated by a nondestructive long-pulsed magnet in the International MegaGauss Science Laboratory in ISSP. Dielectric polarization \(P\) was measured by a pyroelectric technique (Fig. 1). Poling field was typically 200 kV/m. The pyroelectric current was integrated by an analog integrator, and the output voltage was recorded with a 14 bit AD converter (ADC). Typical examples of the data with the top field of 15 T are shown in Figs. 2(a)-(c). The data points were taken with a sampling interval of 5 \(\mu\text{sec}\).

Since the sweep rate of magnetic field \(dB/dt\) is large, \(dP/dt\) and hence the pyroelectric current becomes quite large. Thus a small change in \(P\) can be easily detected. This would be an advantage of the pulsed field measurements. Fig. 2(d) shows a plot of \(P\) versus \(B\) for the data in Figs. 2(a), (c). Large polarization appears in the magnetic field region corresponding to the 5-sublattice-like phase. During the measurements, we noticed that a small spurious voltage in proportion to \(B_{ext}\) was superposed on the data, possibly due to an inductive coupling between the detecting circuit and the time-varying field. We subtracted this spurious component by taking a difference between the data with reversed poling field.

As can be seen in Fig. 2(d), we observed that the polarization amplitude always remains finite when the field has returned to zero. While we do not fully understand the reason, this
may indicate the presence of a finite residual polarization in the sample that would decay slowly with time. This point remains to be clarified in future.

3. Results and Discussions

The field variation of $P$ measured with the top field of 43 T is shown in Fig. 3, together with the magnetization curve obtained for a sample cut out from the same crystal batch. The high-field polarization measurements were done after applying several shots with the top field of 15 T, in order to orient the domains in the 5-sublattice-like phase.

No distinct spontaneous polarization is observed in the newly measured 3-sublattice and 4th phases. Because the 3-sublattice phase takes collinear spin structure, the absence of spontaneous polarization in this phase is consistent with the spin current model. In case of the ideal Heisenberg triangular lattice model, it is expected that the $\sqrt{3} \times \sqrt{3}$ spin structure appears in a whole magnetic field region [12]. In view of the spin current model, even if the spins arrange noncollinearly, the $\sqrt{3} \times \sqrt{3}$ spin structure cancels the magnetically induced microscopic polarization and yields no macroscopic polarization. The magnetization curve suggests that the 4th phase is connected continuously from the 3-sublattice phase which is known to take the $\sqrt{3} \times \sqrt{3}$ spin structure. Thus, the 4th phase might also take the $\sqrt{3} \times \sqrt{3}$ spin structure. Elucidation of the magnetic structure of the 4th phase is desired in the future.

A spontaneous electric polarization of finite size is observed in the 5-sublattice-like phase, in agreement with the previous report [11]. While no hysteresis was reported in [11], we observed a distinct irreversibility in the electric polarization; the spontaneous polarization of the falling field process is evidently smaller than that of the rising field process. This tendency becomes more remarkable when the top field is increased from 15 T to 43 T. We believe that the origin of this behavior is intrinsic, and will discuss in a separate paper. A small step of the polarization is observed between the 5-sublattice and the 3-sublattice phases. As a poling field is reduced, the step becomes smaller suggesting that the tiny change in the electric polarization is due to a difference in the dielectric constant between these two phases.
4. Summary
Dielectric polarization parallel to [110] direction of single crystal of a triangular lattice antiferromagnet CuFeO$_2$ has been measured by pyroelectric technique in pulsed high magnetic field along [001] direction up to 43 T. No distinct spontaneous dielectric polarization is observed in the 4th-field-induced phase with possibly noncollinear spin state and the 4-, 5- and 3-sublattice phase with commensurate collinear spin state. A finite polarization is confirmed only in the 5-sublattice-like phase with incommensurate and noncollinear spin state. The present study demonstrates that the dielectric polarization can be measured in pulsed high magnetic fields.

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