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To cite this version:

HAL Id: jpa-00254793
https://hal.archives-ouvertes.fr/jpa-00254793
Submitted on 1 Jan 1997

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Ferroelectricity of Europium Manganese Oxide EuMn$_2$O$_5$ with Helimagnetic Ordering

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Abstract. From the measurements of pyroelectricity, dielectric constant, magnetic susceptibility and specific heat, we conclude that EuMn$_2$O$_5$ is ferroelectric below the Curie temperature $T_C$ of 38.6 K. This temperature coincides with the Néel temperature of the helimagnetic ordering of Mn$^{3+}$ and Mn$^{4+}$ moments within the accuracy of the measurements. The spontaneous polarization is along the $b$-direction.

1. INTRODUCTION

Rare earth manganites RMn$_2$O$_5$ (R=Nd–Lu, Y or Bi) have a helical magnetic ordering of Mn$^{3+}$ and Mn$^{4+}$ spins below $T_N$ of about 40 K. [1] A strong interrelation is expected between the electric and magnetic properties in these oxides because of the presence of a Mn$^{3+}$ and, in some cases, a rare earth ion which have a tendency to a distorted coordination. In fact, most RMn$_2$O$_5$ show a large magnetoelectric effect at low temperatures.[2] Particularly, we proposed the possibility of a simultaneous ferroelectric transition at $T_N$ in EuMn$_2$O$_5$, based on the dielectric, magnetic and magnetoelectric measurements.[3] However, direct demonstration of ferroelectricity has been lacking. The purpose of this study is to make this point clear. We made pyroelectric measurements to demonstrate the presence of spontaneous polarization and determine its temperature dependence. In addition, we made simultaneous measurements of dielectric constant and magnetic susceptibility, and specific heat measurements in order to know whether both ferroelectric and magnetic transitions take place at the same temperature. The results is that EuMn$_2$O$_5$ is ferroelectric below 40 K, which coincides with magnetic transition temperature within ±0.3 K.

2. EXPERIMENTAL METHODS

We used two kinds of samples. The sample for the measurement of pyroelectricity is a single crystal of the same batch of the one used in a previous study.[3] The others were polycrystals prepared by an ordinary ceramic method. The composition determined by ICP analysis was Eu$_{1.00}$Mn$_{2.02}$O$_{5.02}$. No line of phases other than EuMn$_2$O$_5$ could be observed in the powder X-ray pattern, though the small remanent magnetization indicated the presence of the precipitate of Mn$_3$O$_4$ or EuMnO$_3$. The polycrystalline samples were used in the measurements of the dielectric constant, magnetic susceptibility and specific heat. The pyroelectric current was detected along the $b$-direction of the single crystal sample with an electrometer. Preceding to each measurement run, the samples were poled by cooling down from 77 to 4.2 K in electric field of 5 or -3 kV/cm along the $b$-direction. The spontaneous polarization $P_S$ was derived by integrating the recorded current. The details were described elsewhere.[4] The magnetic susceptibility was measured by a vibration sample magnetometer with a specially designed sample rod. The dielectric constant was measured simultaneously by an impedance analyzer connected to the electrodes on the sample through coaxial cables set in the sample rod. Specific heat measurements were carried out with a heat-pulse and a continuous heating method in an adiabatic calorimeter installed at the Institute for Solid State Physics, University of Tokyo. The relative calibration of the temperature scales of two kinds of measurements were not carried out yet.

3. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the spontaneous polarization $P_S$ along the $b$-direction. With increasing temperature, $P_S$ decreases monotonously and vanishes at about 39 K. Two curves correspond to opposite directions of electric field $E_{cool}$ applied during cooling the sample down to 4.2 K. The sign of $P_S$ was switched by reversing $E_{cool}$. Figure 2 and Figure 3 depict the temperature dependencies of the magnetic susceptibility and dielectric constant measured simultaneously. An arrow indicates the temperature $T_N$ of the magnetic transition estimated in the following way; we reconstructed the
temperature dependence of powder susceptibility $\chi_p$ from our previous data of the susceptibilities $\chi_x$, $\chi_y$, and $\chi_z$ along the three principal axes of a single crystal (shown in the inset of Fig. 2) [3], and then we determined the point corresponding to $T_c$ by comparing the recorded and the reconstructed temperature dependence of $\chi_p$ on an assumption that the cusp in $\chi_p$ corresponds with $T_c$. The accuracy of the determination was within 0.3 K. Figure 4 shows the temperature dependence of specific heat. Only a single peak is present around 40 K (see the inset for more detail), showing again that both ferroelectric and magnetic transitions occur in a narrow temperature range.

The above results indicate that EuMn$_2$O$_5$ has spontaneous polarization, which can be switched by external electric field, below $T_c$ of 40 K. In other words it is ferroelectric below this temperature. The ferroelectric Curic temperature $T_c$ and the Néel temperature $T_N$ are coincident within 0.3 K. If the formation of magnetic ordering is accompanied with ionic displacement leading to ferroelectricity, it is a new type of transition. The mechanism is not yet clear. However, it is probable that the magnetic long range ordering brings the change in the state of a Mn$^{3+}$ ion with 3d$^4$ configuration and then the relative displacement of it relative to surrounding oxygen ions. It is a subject of our further study.

References