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Ferroelectricity of Europium Manganese Oxide EuMn_2O_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_2O_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_2O_5 ($R=\text{Nd-Lu, Y or Bi}$) 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_2O_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_2O_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_2O_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 $\text{Eu}_{1.00}\text{Mn}_{2.02}\text{O}_{5\pm\alpha}$. No line of phases other than EuMn_2O_5 could be observed in the powder X-ray pattern, though the small remanent magnetization indicated the presence of the precipitate of Mn_3O_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 χ_p from our previous data of the susceptibilities χ_a , χ_b and χ_c 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_N by comparing the recorded and the reconstructed temperature dependence of χ_p on an assumption that the cusp in χ_a corresponds with T_N . 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_2O_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 Curie 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

Figure 1: Temperature dependence of spontaneous polarization of EuMn_2O_5 along the b -direction. Two curves correspond to opposite direction of electric field applied during cooling.

Figure 3: Temperature dependence of dielectric constant of powder sample of EuMn_2O_5 . The data of Figures 2 and 3 were recorded in the same experimental run.

Figure 2: Temperature dependence of the observed (triangles) and the estimated (circles) powder magnetic susceptibility of EuMn_2O_5 . The inset shows the temperature dependence of principal susceptibilities used in the estimation.

Figure 4: Temperature dependence of specific heat of EuMn_2O_5 . The inset shows the data measured between 38 and 43 K with a continuous heating method.

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.

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