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Dielectric and Magnetic Properties of $BaTiO_3$-$LaMnO_3$ Composites

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We have investigated the dielectric and magnetic properties of ferroelectric-antiferromagnetic $BaTiO_3$-$LaMnO_3$ composite with changing relative mole percents. Due to high sintering temperature, i.e. 1150 °C, the Ba ion in $BaTiO_3$ seems to diffuse into $LaMnO_3$; resulting in $BaTiO_3$-(La,Ba)MnO_3 ferroelectric-ferromagnetic composite. At room temperature, 0.9$BaTiO_3$-0.1$LaMnO_3$ composite exhibits considerable magnetization (~0.7 emu/g at 2000 Oe) and low coercive field (~5 Oe). Also it exhibits high dielectric constant (~560) and low loss (~0.08) at 10 kHz. This result may imply that $BaTiO_3$-$LaMnO_3$ could be suitable for a low leakage multiferroic composite.

Keywords : multiferroic, $BaTiO_3$-$LaMnO_3$, ferroelectric-ferromagnetic composite, low leakage

1. Introduction

In recent years, lots of attentions have been paid on multiferroic materials, which exhibit (anti)ferromagnetic and (anti)ferroelectric properties simultaneously [1]. Due to a strong coupling between magnetism and electricity, multiferroics show a novel magnetoelectric phenomenon, i.e., the control of magnetization and polarization (dielectric constant) by applying external electric and magnetic fields, respectively [2].

However, most single phase multiferroics have quite small electric polarization and/or magnetization, and the coexistence of (anti)ferromagnetic and (anti)ferroelectric properties occurs at quite low temperature [3]. Although the magnetoelectric coupling is quite strong, it is quite difficult to use them for application. On the other hand, several composite multiferroics show large electric polarization and magnetization even at room temperature [4]. However, the magnetoelectric coupling is quite small and the composite samples usually show quite large loss at and above room temperature.

Therefore, it would be quite useful if we explore a composite which has large electric polarization and magnetization with small loss at room temperature. In this paper, we report our on-going research for dielectric and magnetic properties of (1-x)$BaTiO_3$-x$LaMnO_3$ composites at room temperature. Near x~0.1, it shows ferromagnetism with small dielectric loss. Our work may suggest a new approach for the design of multiferroics, composed of two compounds with same crystal structures.

2. Experiments

A series of $BaTiO_3$-$LaMnO_3$ composites were synthesized by a standard solid-state reaction method. $BaTiO_3$ and $LaMnO_3$ pellets were separately prepared by the conventional calcination and sintering processes. The $BaTiO_3$ and $LaMnO_3$ pellets were crushed into powders and mixed with appropriate mole percents. The mixed powders were pressed into pellets under 500 MPa by using a cold isostatic press. The mixed pellets were sintered at 1150 °C for 24 hrs with flowing of oxygen gas.

Magnetic property of composites was examined by a superconducting quantum interference device magnetometer. And, complex dielectric constant was evaluated by an LCR meter with an excitation of 1 V. For dielectric measurement, the samples were cut into the form of a thin plate of thickness ~0.5 mm, and gold electrodes of area ~14 mm^2 were evaporated on both sides of the samples.

3. Results and Discussion

Figure 1 shows the x-ray diffraction patterns of (1-x)$BaTiO_3$-x$LaMnO_3$ composites around the (200) peak of $BaTiO_3$. [Wide scan x-ray diffraction pattern (not shown) reveals no other crystalline phases except perovskite ones.] Due to the tetragonal structure of $BaTiO_3$, the (200)

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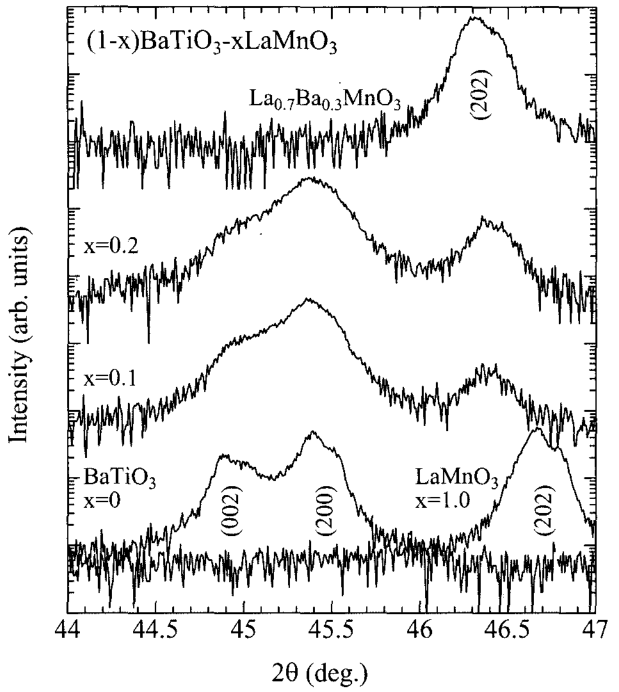

Fig. 1. X-ray diffraction patterns of (1-x)$BaTiO_3$-x$LaMnO_3$ composites. For comparison, we also show the pattern of $La_{0.7}Ba_{0.3}MnO_3$.

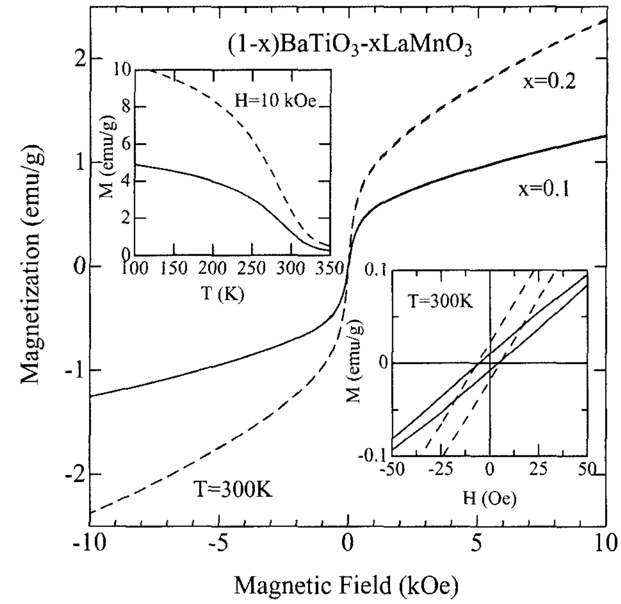

Fig. 2. Magnetic field dependent magnetization at room temperature for x = 0.1 and 0.2 of (1-x)$BaTiO_3$-x$LaMnO_3$ composites. In upper and lower insets, we also show the temperature dependent magnetization and magnified magnetic field dependent magnetization, respectively.

peak is split into (002) and (200) peaks. It should be noted that the tetragonal structure of $BaTiO_3$ is originated from the relative movement of Ti ion to O one in perovskite structure and quite crucial for its ferroelectricity [5]. With increasing x, the (002) peak intensity starts to decrease. However, one may notice a shoulder peak at the same (002) peak position in addition to (200) peak even at x = 0.2; which might imply that the ferroelectricity remains in this composite. On the other hand, instead of $LaMnO_3$ (202) peak near 46.7°, a new peak near 46.3° show up in all $BaTiO_3$-$LaMnO_3$ composites. To identify the new peak, we show the x-ray diffraction pattern of $La_{0.7}Ba_{0.3}MnO_3$. Interestingly, the new peak near 46.3° is consistent with (202) peak in $La_{0.7}Ba_{0.3}MnO_3$. This result implies that due to high sintering temperature, the Ba ion in $BaTiO_3$ diffuse into $LaMnO_3$, which results in (La,Ba)MnO_3 [6]. Note that $La_{1-y}Ba_yMnO_3$ shows quite complicate electrical and magnetic properties depending on y and temperature. At room temperature, especially, it is paramagnetic insulator for $0 \leq y \leq 0.2$ and ferromagnetic metal for $0.2 < y < 0.5$ [7].

In the upper inset of Fig. 2, we show the temperature dependent magnetization for x=0.1 and 0.2 in (1-x)-$BaTiO_3$-x$LaMnO_3$ composite. Clearly, both magnetization values increase with decreasing temperature. By the inflection point of magnetization, we estimate the Curie temperature of x = 0.1 and 0.2 at 315 K and 318 K, respectively. It suggests that both composites are ferromagnet at room temperature.

Figure 2 shows the magnetic field dependent magnetization curves of x = 0.1 and 0.2 at room temperature. Both composites show ferromagnetic hysteresis behavior. However, they do not show saturated magnetization but keep increasing even at 10 kOe. From the magnified hysteresis curves (the lower inset), we estimate coercive field as 5 Oe for both x = 0.1 and 0.2. The values of the Curie temperature and coercive field are similar to the reported $La_{1-y}Ba_yMnO_3$ (y~0.3) [8].

For the possible application of $BaTiO_3$-$LaMnO_3$ composite as a multiferroic, one of the important physical parameters is a complex dielectric constant ($=\varepsilon_1 + i\varepsilon_2$). In Figs. 3(a) and 3(b), we show the frequency dependent dielectric constant (ε_1) and loss $\tan\delta(=\varepsilon_2/\varepsilon_1)$, respectively, for the frequency region from 100 Hz to 100 kHz.

Similar to previous reports, our $BaTiO_3$ (x = 0) shows very large dielectric constant (~1800) with small frequency dependence, and quite small loss (~0.02) [9]. [Note that, the value of the dielectric constant of $BaTiO_3$ is scaled down by factor of two for clarity.] In x = 0.1 and 0.2, however, the dielectric constants become reduced by factor of two and show strong frequency dependences. Also, their dielectric losses start to increase, especially below 10 kHz.

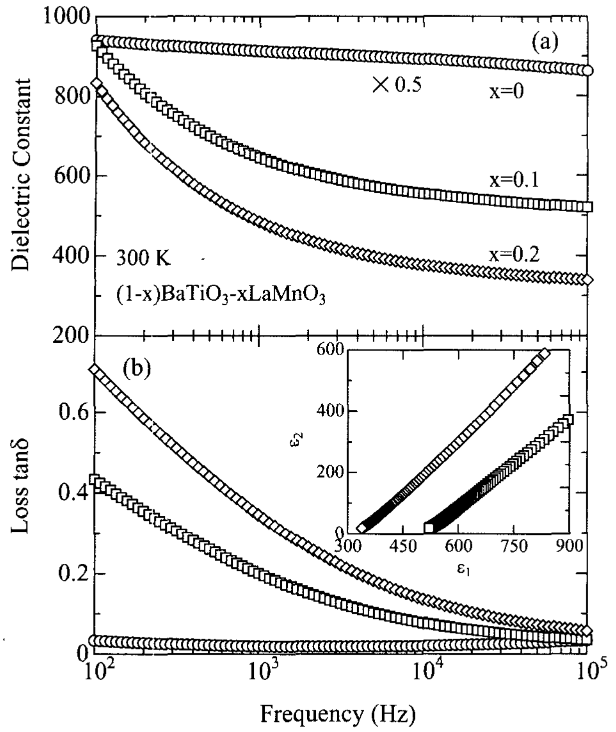

Fig. 3. Frequency dependent (a) dielectric constant and (b) loss tanδ for x = 0 (open circles), 0.1 (open squares), and 0.2 (open diamonds) of (1-x)$BaTiO_3$-x$LaMnO_3$ composites. In the inset (b), Cole-Cole plots are shown for x = 0.1 and 0.2.

To investigate the relation between dielectric constant and loss, in the inset of Fig. 3(b), we show a Cole-Cole plot, i.e., the plot between ε_1 and ε_2. It is well known that if there is quite small loss and only one relaxation, the Cole-Cole plot should be a semi-circular. If there is large loss or several relaxations, on the other hand, the semi-circle becomes distorted and a linear line [10]. In our Cole-Cole plots for x = 0.1 and 0.2, one may clearly notice the linear behaviors. We believe that the loss in our composites originates from a space charge in $BaTiO_3$-(La,Ba)MnO_3 interfaces and/or the presence of metallic (La,Ba)MnO_3 phase, similar to other insulator/conductor composites [11]. However, one should note that the values of loss tanδ ~0.1 at 10 kHz in our (1-x)$BaTiO_3$-x$LaMnO_3$ (x=0.1 and 0.2) composites are quite smaller than $CdCr_2S_4$ single crystal [12] but close to those in recent $BaTiO_3$-$Ni_{0.5}Zn_{0.5}Fe_2O_4$ composite [11].

4. Conclusion

The dielectric and magnetic properties of $BaTiO_3$-$LaMnO_3$ composites reveal a low loss ferromagnet at room temperature. The magnetoelectric coupling in this composite is under measurement. Our result may imply that several multiferroic composites, such as $PbTiO_3$-$LaMnO_3$, with possibly low loss and high magnetization at room temperature may be achieved by the simple diffusion of ion at appropriate temperature.

Acknowledgments

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