



Magnetoelectric properties of multiferroic composites $(1-x)$ ErMnO_3 – $x\text{Y}_3\text{Fe}_5\text{O}_{12}$ at room temperature



B. Raneesh^{a,b}, H. Soumya^c, J. Philip^c, S. Thomas^b, K. Nandakumar^{a,b,*}

^a School of Pure and Applied Physics, Mahatma Gandhi University, Kottayam, Kerala 686 560, India

^b International and Inter University Centre for Nanoscience and Nanotechnology, Mahatma Gandhi University, Kottayam, Kerala 686 560, India

^c Sophisticated Test and Instrumentation Centre, Cochin University of Science and Technology, Cochin 682 022, India

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ABSTRACT

We report multiferroic behavior of the composites $(1-x)\text{ErMnO}_3$ – $x\text{Y}_3\text{Fe}_5\text{O}_{12}$ ($0 \leq x \leq 1$) with spin–charge coupling at room temperature. These composites have been synthesized by sol–gel technique followed by solid-state reaction. Crystalline phases and microstructures of the composites are examined using X-ray diffraction (XRD) and transmission electron microscope (TEM) techniques. The XRD results indicate that the composites consist of both hexagonal ErMnO_3 and cubic $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG) phases. Dielectric constant decreases with increase in YIG concentration. Magnetic properties of the composite samples measured as a function of YIG concentration show soft magnetic behavior, with magnetization increasing with increasing YIG content. Strong magnetoelectric (ME) coupling is demonstrated in the composite 0.5ErMnO_3 – 0.5YIG by a dynamic lock-in amplifier set up. Thermal measurements using photopyroelectric technique indicate that thermal conductivity decreases with increase in YIG content or phonon transport through these composites is effectively suppressed with YIG concentration. These results provide data and criteria for the design and fabrication of potential devices based on this room temperature multiferroic material.

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1. Introduction

Recently, there has been a revival of interest in multiferroic composites in which magnetoelectric (ME) coupling takes place indirectly via strain. Commonly, ferroelectrics in combination with ferromagnets are used for this purpose [1–3]. The coupling coefficients achieved in these materials is often many orders of magnitude larger than in single-phase multiferroics. The compound ErMnO_3 is a well-known example for a single phase multiferroic whose peculiar properties are due to a combination of magnetic frustration and magneto-elastic coupling. Electrical properties of ErMnO_3 are quite interesting because of the presence of acentric structures. Unlike major classes of ferroelectric materials these materials exhibit electronically driven distortions due to geometric packing effects [4–6]. In general, ME effect in most single phase multiferroic materials exists only at very low temperatures [7,8]. Many micromechanical models have been suggested for two-phase composites envisaging product of moduli in multiferroic

composites [9–13]. In this work, Yttrium iron garnet (YIG) has been selected as the magnetic counterpart due to its interesting magneto-optical properties and ErMnO_3 as the electrical counterpart due its intrinsic multiferroic properties at low temperatures. This could lead to a novel structure and may provide new way to investigate the complex interfacial charge–spin interactions.

YIG is a well-known magneto-optical medium, which is almost transparent to the visible region above 500 nm [14]. The Curie temperature of this compound is approximately 555 K. The fundamental absorption edge determined by the charge transfer transition is 2.85 eV [15,16]. The crystal lattice of the compound has two nonequivalent (octa and tetrahedral) positions occupied by Fe^{3+} ions in the ratio of 2:3. The magneto-optical effect of garnet medium gives an additional degree of freedom with ErMnO_3 structures. However, there are no reports on the synthesis and characterization of ErMnO_3 –YIG composite multiferroics till now. Problems associated with the preparation of this type of multiferroic composites could be due to inter-diffusion and thermal mismatch between the piezoelectric and magnetic phases, which are still very much unclear. In order to explain this behavior, detailed experimental evidences are needed. In this paper, we report the results of investigations carried out on the structural, dielectric,

* Corresponding author at: School of Pure and Applied Physics, Mahatma Gandhi University, Kottayam, Kerala 686 560, India. Tel.: +91 9447671962; fax: +91 481 2731669.

E-mail address: nkkalarikkal@mgu.ac.in (K. Nandakumar).

magnetic and thermal properties of ErMnO_3 –YIG composites synthesized by sol–gel method, followed by solid state reaction.

2. Experimental

Ceramic composites of $(1-x)\text{ErMnO}_3$ – $x\text{Y}_3\text{Fe}_5\text{O}_{12}$, with different molar weight fractions ($x = 0.1, 0.3, 0.5, 0.7$ and 0.9), were prepared by sol–gel technique followed by solid state reaction method. First, $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG) and ErMnO_3 nanoparticles were synthesized by a sol–gel method as described elsewhere [16,17]. In order to synthesize the composite, YIG and ErMnO_3 powders were mixed thoroughly in appropriate stoichiometric proportions and sintered at 750°C for 5 h after grinding them for 6 h. The low sintering temperature of the composites helped to prevent abnormal grain growth in the system. The sintered composites exhibited several advantages such as freedom in the selection of constituent phases, their starting particle sizes, processing parameters etc. Structural and morphological studies were carried out by powder X-ray diffraction (XRD, PANalytical X'Pert Pro) and Transmission electron microscopy (TEM, Jeol JEM-2100). Energy Dispersive X-ray analysis (EDX) was used to check the overall chemical homogeneity and composition of the samples. The magnetic properties were measured with a vibrating sample magnetometer (VSM, LDJ 9600-1) at room temperature. Dielectric measurements were performed using an Impedance analyzer (Agilent, E4980A). A dynamic lock-in amplifier technique was used to measure the direct ME coupling coefficients of composites. The thermal conductivities of the samples were measured using a photopyroelectric (PPE) thermal wave technique. For this measurement, the sample thickness should be such that the sample, the pyroelectric detector and the backing material used were thermally thick in the frequency range used in the measurement [18,19].

3. Results and discussion

Fig. 1 shows the powder XRD patterns of the multiferroic composites $(1-x)\text{ErMnO}_3$ – $x\text{Y}_3\text{Fe}_5\text{O}_{12}$ ($x = 0, 0.1, 0.3, 0.5, 0.7, 0.9$ and 1) which consist of two major types of phases; hexagonal ErMnO_3 and cubic $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG). It shows that intensity of YIG peaks become stronger with the increase of YIG content in the composites. Moreover, in the X-ray diffraction profiles, no additional peaks from impurity phases could be detected. All of the diffraction peaks can be indexed with reference to the standard powder diffraction patterns of ErMnO_3 and $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (JCPDS Nos.14-0689 and 43-0507). The phase analysis shows the formation of cubic–hexagonal mixed structure in the composites.

The low-resolution TEM image of $(1-x)\text{ErMnO}_3$ – $x\text{Y}_3\text{Fe}_5\text{O}_{12}$ composites with 50–50 M fraction is shown in Fig. 2(a). The two crystalline phases of the fine-grained and dense microstructure ceramic composite of YIG and ErMnO_3 grains are also confirmed by the TEM images. Large thermal expansion mismatch between the ferroelectric and ferrite phases harms the densification and leads to the formation of microcracks in the systems. Fig. 2(b) shows the high resolution TEM (HRTEM) image of a 0.5ErMnO_3 – $0.5\text{Y}_3\text{Fe}_5\text{O}_{12}$, which indicates 0.272 nm spacing between two

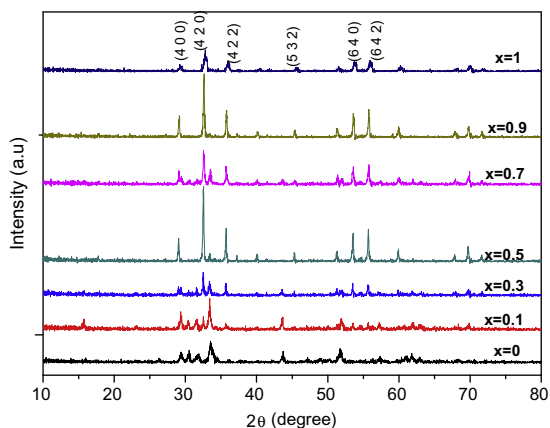


Fig. 1. Powder X-ray diffraction patterns of the composites $(1-x)\text{ErMnO}_3$ – $x\text{Y}_3\text{Fe}_5\text{O}_{12}$ ($0 \leq x \leq 1$).

adjacent lattice planes corresponding to the (101) lattice planes of ErMnO_3 , and a spacing 0.431 nm can be identified for the (220) planes of $\text{Y}_3\text{Fe}_5\text{O}_{12}$ phase. Clear lattice fringes propose that the samples are defect free and highly crystalline in nature. The HRTEM image also shows a clear interface between ErMnO_3 phase and $\text{Y}_3\text{Fe}_5\text{O}_{12}$ phase. Movement of strain between the ferrite and ferroelectric phases could happen at this interface and it might be favourable to build a good ME coupling in the system. A typical energy dispersive X-ray (EDX) spectrum of 0.5ErMnO_3 – $0.5\text{Y}_3\text{Fe}_5\text{O}_{12}$ is shown in Fig. 2(c). The elemental analysis of the composites confirms the presence of elements Er, Y, Mn, Fe, and O (Cu is due to the TEM copper grid). Moreover, the EDX analysis indicates that the compositions are nearly stoichiometric.

The dielectric properties of multiferroic $(1-x)\text{ErMnO}_3$ – $x\text{YIG}$ have been determined over a broad frequency range between 100 Hz and 2 MHz at room temperature. The effective dielectric properties of the multiferroic composites exhibit strong frequency dependence, where the dielectric constant (ϵ') and loss factor ($\tan \delta$) show a decrease in their values as frequency increases from 100 Hz to 2 MHz, as shown in Fig. 3. The high value of dielectric constant observed at low frequencies is explained on the basis of space charge polarization due to inhomogeneous dielectric structure and Maxwell–Wagner type interfacial polarization [20,21], which is in agreement with Koops phenomenological theory [22,23]. The frequency dependences of the dielectric properties for different compositions show that the inclusion of YIG has a significant effect on dielectric properties due to the different microstructures of YIG in the ErMnO_3 structure. The dielectric constant of the composites initially increases with YIG concentration up to 30 mol% and then decreases. However, the inhomogeneous crystalline nature of the composites gives rise to an additional interfacial polarization. The high value of dielectric constant is ascribed to the fact that ferroelectric regions are surrounded by ferrite regions. As the ferroelectric content decreases (say, mol fraction from $x = 0.50$ to 0.90), dielectric constant is found to decrease showing the dependence of dielectric property on ErMnO_3 content present in the samples. The increase of ferrimagnetic phase decreases the number of dipoles contributing to dielectric polarization. The value of $\tan \delta$ measures the loss of electrical energy from the applied electric field into the samples at different frequencies (Fig. 3(b)). None of the samples exhibit any loss peak. The low loss values at higher frequencies show the potential applications of these materials in high frequency microwave devices. The dielectric properties of ErMnO_3 –YIG composite samples are due to the presence of mutual interactions between the ferroelectric and ferrimagnetic orders and require further studies. For further understanding, the studies of the complex permeability and permittivity spectra of composite materials as a function of temperature need to be performed.

In an effort to understand the magnetic properties of these composites better, vibration sample magnetometer (VSM) measurements were performed on the samples at room temperature. Fig. 4(a) shows the variations of magnetization (M) with applied field (H) for the $(1-x)\text{ErMnO}_3$ – $x\text{YIG}$ composites. All the composites exhibit ferrimagnetic behavior with a saturation magnetization (M_s) and the value of the saturation magnetization increases with the concentration of the YIG. The results indicate hysteresis in the field range ± 400 Oe, while outside this range the specific magnetization increases with increasing field and saturates at high field. All the samples of $(1-x)\text{ErMnO}_3$ – $x\text{YIG}$ show the same features for saturation magnetization, which increases from 11 to 48 emu/g with increase in YIG concentration from $x = 0$ to 0.9 (Fig. 4(a)). The coercivity (H_c) also rises with the increase of YIG concentration. The minimum and maximum values of coercivity are found to be 37 Oe and 83 Oe respectively (Fig. 4(b)). Materials with very low intrinsic coercivity are good soft magnetic materials and the

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