Research Article

MAGNETOELECTRIC EFFECTS IN FERROMAGNETIC/PIEZOELECTRIC MULTILAYER COMPOSITES

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ABSTRACT:

The observation of strong magnetoelectric effects is reported in thick film bilayers and multilayers of ferrite-lead titanate zirconate (PZT) and lanthanum nanganite-PZT. The ferrites used in our studies included pure and zinc substituted cobalt-, nickel- and lithium ferrites. Samples were prepared by sintering 10-40 µm thick films obtained by tape-casting. Measurements of ME voltage coefficients at 10-1000 Hz indicated a giant ME effect in nickel ferrite-PZT, but a relatively weak coupling in other ferrite-PZT and manganite-PZT systems. Multilayers prepared by hot pressing was found to show a higher ME coefficient than sintered samples. Evidence was found for enhancement in ME coefficients when Zn was substituted in ferrites. The Zn-assisted increase was attributed to low anisotropy and high permeability that resulted in favorable magneto-mechanical coupling in the composites. We analyzed the data in terms of our recent comprehensive theory that takes into account actual interface conditions by introducing an interface coupling parameter. Theoretical longitudinal and transverse ME voltage coefficients for unclamped and clamped samples are in general agreement with data. From the analysis we inferred excellent interface coupling for nickel zinc ferrite-PZT and weak coupling for other layered systems.

Keywords: HPLC, PDA, stability indication method, drug.

1. INTRODUCTION:

This work is concerned with the fabrication of novel ferromagnetic-ferroelectric thick film multilayers and studies on the nature of magnetoelectric interactions. In such two-phase composites, the magnetoelectric (ME) coupling is mediated by mechanical stress [1]. An applied ac magnetic field produces dynamic deformation in ferromagnets due to magnetostriction and results in an induced electric field due to piezoelectric effect. The systems of interest in the past were bulk samples of nickel or cobalt ferrite with BaTiO3 or lead zirconate titanate (PZT) that showed ME coupling much smaller than predicted values [2-5]. The main cause is low resistivity for ferrites that gives rise to a leakage current and limits the electric field for orienting the dipoles, leading to loss of charges and poor piezoelectric coupling. Such problems could be eliminated in a layered structure [6]. We recently initiated studies on layered heterostructures. The main emphasis of our work has been on ferromagnetic manganite-PZT and ferritePZT. Samples were synthesized by sintering thick films made by tape casting and characterized by structural, magnetic, and electrical measurements. Magnetoelectric measurements were made both at low (10 Hz–1 kHz) and high frequencies (9-10 GHz). Key findings and accomplishments are as follows. (i) The first observation of ME coupling in lanthanum strontium manganite-PZT [7]. (ii) A giant low frequency ME interaction in ferrite-PZT [8-11]. (iii) Ultrahigh ME effects at electromechanical resonance (EMR). (iv) Analysis of low frequency ME data using a model for a bilayer that allows the estimation of an all-important interface coupling constant [12,13]. (v) Theoretical models for resonance ME coupling: at EMR for the piezoelectric phase and ferromagnetic resonance for the ferromagnetic phase [14-16]. Our efforts so far have resulted in considerable progress toward an understanding of ME interactions in layered systems [8-19]. The composites are candidate materials for magnetoelectric memory devices, smart sensors, and electric or magnetic field controlled signal-processing devices.

2. RELATED STUDY:

Materials with features on the scale of nanometer often have properties dramatically different from their bulk scale counterparts. Nanocrystalline materials are single phase or multiphase polycrystals, the crystal size of which is of the order of few nanometers so that about 40 to 80 % of the atoms are in the grain boundaries [1]. Nanostructure science and technology is a broad and interdisciplinary area of research and development activity that has been growing worldwide in the past decades [2]. Important among these nanoscale materials are nanocomposites, in which the constituents are mixed at nanometer length scale. They often have properties that are different compared to conventional microscale composites and can be synthesized using simple and inexpensive techniques. The study of nanocomposite materials requires a multidisciplinary approach with impressive technological promise, involving novel synthesis techniques and an understanding of physics and surface science [3]. During the last decade, the development of magnetic nanocomposite materials has been the source of discovery of spectacular new phenomena, with potential applications in the fields of information technology, telecommunication or medicine [4, 5]. Magnetic nanocomposite materials are generally composed of ferromagnetic particles (grain size in nanometer scale) distributed either in a non-magnetic or magnetic matrix [6, 7]. The shape, size and distribution of the magnetic particles play an important role in determining the properties of such materials [8]. The matrix phase separates the magnetic particles and changes the magnetic exchange interaction. This affects the transport and magnetic properties. Therefore, understanding and controlling the structure of materials is essential to obtain desired physical properties. Hence in this chapter, a general introduction to the important magnetic properties of nanomaterials, colossal magnetoresistance (CMR) materials, and various electrical transport mechanisms are described based on literature. The organization of thesis is given in the last part of this chapter. The main objective of the present work is presented at the end of the second chapter, which is based on a critical literature survey.

3. PROPOSED METHODOLOGY:

The effective magnetic moment of a ferromagnetic particle is determined by its size. A ferromagnetic sample with a volume greater than a critical value Vc divides into multiple magnetic domains, each magnetized along the local easy axis but in one of two opposite Fig. 1.1: Overview of the size dependence of coercivity exhibited by magnetic particles: HC = 0 below superparamagnetic (SP) particle size limit r0; single-domain behavior (SD) between r0 and the single domain limit rc; and multidomain behavior (MD) for r > rc. [Adapted from ref. 9] 4 directions. The multiple domain structure is, however, no longer favorable below the critical volume, and the particle becomes a single domain with ferromagnetic alignment of all its moments along the easy axis in the same direction. Thermal fluctuations of the moment exist on a microscopic scale, but to reverse the direction of the single domain's magnetization requires an energy ΔE to overcome the crystal-field anisotropy. If single domain particles become small enough, KV would become so small that thermal fluctuations could overcome the anisotropy forces and spontaneously reverse the magnetization of a particle from one easy direction to the other, even in the absence of an applied field. Each particle has a magnetic moment μ = MsV and, if a field is applied, the field will tend to align the moments of the particles and the thermal energy will tend to disalign them. This is called superparamagnetism. The probability of such a reversal by thermal activation is proportional to exp (-ΔE/kT). This differs from conventional paramagnetism because the effective moment of the particle is the sum of its ionic particles, which can be several thousand spins in a ferromagnetic particle small enough to show superparamagnetism [9]. Very fine ferromagnetic particles have very short relaxation times even at room temperature and behave superparamagnetically; that is, their behavior is paramagnetic but their magnetization values are typical of ferromagnetic substances. The individual particles have normal ferromagnetic moments but very short relaxation times so that they can rapidly follow directional changes of an applied field and, on removal of the field, do not hold any remanent moment. Superparamagnetism is characterized by two experimental features: 1. There is no hysteresis; (i.e., both the retentivity and the coercivity are zero) in the field dependence of magnetization. 2. Magnetization curves measured at different temperatures superimpose when magnetization (M) is plotted as a function of Field (H) / temperature (T). Superparamagnetism can be destroyed by cooling. This follows because the characteristic fluctuation time for a particle's moment varies exponentially with temperature, so the magnetization appears to switch sharply to a stable state as the temperature is reduced. The temperature at which this occurs is called the blocking temperature (TB), and it depends linearly on the sample's volume and on the magnitude of the crystal-field anisotropy. In the case of superparamagnetic materials, the magnetization shows temperature and path dependence which is shown schematically. The two curves zero field cooled (ZFC) and field cooled (FC) show different behavior at low temperatures. As the temperature increases the magnetic moment in the FC curve decreases. However, as the temperature begins to rise from 5 K, the moment in the ZFC curve begins to increase. At a certain temperature, the ZFC curve reaches a peak and this temperature is called the blocking temperature (TB). The divergence of ZFC and FC curve and the blocking temperature depend on the particle size and its distribution. The blocking temperature of a substance should decrease with increasing applied field and eventually disappear when the field reaches a critical value. The higher field is expected to lower the barriers between the two easy axis orientations.

Our efforts have been mainly on bilayers and multilayers (MLs) of lanthanum manganites-PZT and ferrite-PZT [8-14]. Layered composites were synthesized using thick films of manganites, ferrites and PZT obtained by tape-casting [27]. The ferrite/manganite powder necessary for tape casting was prepared by the standard ceramic techniques that involved mixing the oxides or carbonates of the constituent metals, followed by presintering and final sintering. A ball-mill was used to grind the powder to submicron size. For PZT films, we used commercially available powder. The fabrication of thick films contained the following main steps: a) preparation of cast of constituent oxides; b) deposition of 10-40 µm thick films tapes by doctor blade techniques; and c) lamination and sintering of composites. Ferrite or PZT powders were mixed with a solvent (ethyl alcohol), plasticizer (butyl benzyl phthalate), and binder (polyvinyl butyral) in a ball mill for 24 hrs. The slurries were cast into 10-40 µm tapes on silicon coated mylar sheets using a tape caster. The films were dried in air for 24 hrs, removed from the mylar substrate and arranged to obtain the desired structure. They were then laminated under high pressure (3000 psi) and high temperature (400 K), and sintered at 1375-1475 K. Bilayers were made with 200 µm thick ferrite or manganite and PZT. Multilayers consisted of (n+1) layers of ferrites/manganite and n layers of PZT (n = 5-30).

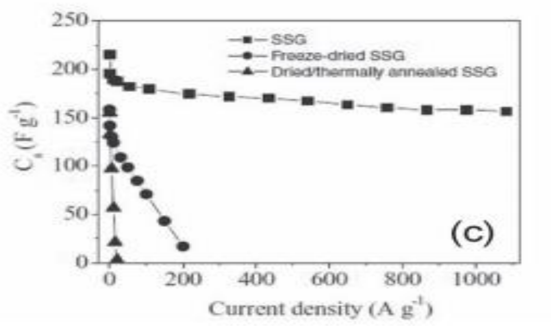

Fig.4.1. Gravimetric capacitances measured at various charge/discharge currents.

5. CONCLUSION:

Studies on layered samples of lanthanum manganite-PZT and pure and zinc substituted ferrites - PZT show evidence for strong ME interactions. Of particular interest is the giant ME voltage coefficients in nickel ferrite-PZT. The voltage coefficient αE show an overall increase with increasing Zn concentration x in CZFO-PZT and NZFO-PZT. A maximum in αE occurs for x = 0.2-0.4, depending on the ferrite. Analysis of the data using our model for a bilayer reveals ideal interface conditions for NZFOPZT. The data implies poor coupling in other ferritePZT and manganite-PZT. The Zn assisted enhancement in ME coefficient is primarily due to low anisotropy and high permeability for the ferrites that result in favorable magneto-mechanical coupling in the composites.

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