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Room Temperature Magnetoelectric Coupling and AC Electrical Response of 0.2 $NiFe_2O_4$J 0.8 ZnO Multiferroic Nanocomposite

Papia Dutta
Department of Physics, National Institute of Technology Agartala, Agartala, Tripura , India
S. K. Mandal
Department of Physics, National Institute of Technology Agartala, Agartala, Tripura , India
A. Nath
Department of Physics, National Institute of Technology Agartala, Agartala, Tripura , India

ABSTRACT

Multiferroic 0.2$NiFe_2O_4$ – 0.8 ZnO nanocompositeshave been prepared through low temperature chemical "pyrophoric reaction process" followed by solid state route. Structural characterization has been carried out through X-ray diffraction technique,which shows the co-existence of both the phases of the nanocomposites. The ac electrical properties of nanocomposites have been studied employing impedance spectroscopy technique. The impedance value is found to increase with increase in magnetic field attributing the magnetostriction property of the composites. Dielectric constant is found to decrease with increase in magnetic field. The magnetoelectric voltage has been obtained for both longitudinal and transverse direction. The value of magnetoimpedance and magnetodielectric is found to ~43 and ~ 34 % for magnetostriction property of the magnetostrictive materials.

Keywords

Nanocomposites, Impedance Spectroscopy, Dielectric, Magnetoelectric Coupling.

INTRODUCTION

Magnetoelectricmultiferroics presenting coupled magnetic and ferroelectric order parameters have attracted a lot of attention towards the design of new materials to outfit the desired application. In semiconductor nanocrystal, there is an increasing research interest considering the issue of size miniaturization and surface state on nanometer scale [1]. Zinc Oxide (ZnO), hexagonal wurtzite structure, which is well known for its wide band gap of 3.37 eV and large excitonic binding energy of 60 meV, has been of growing technological interest due to its versatile favourable properties, achieved great variety of applications such as in data storage devices, photodetector, magneto-electric based devices, sensors or photocatalysis [2,3]. In particular, ZnO holding an interesting and preferential candidate for functioning as a multipurpose material that combines superior optical, chemical, electrical, piezoelectric and magnetic properties. Nowadays, multiferroic material constituting ferromagnetic and ferroelectric properties is a subject of intense research interest. Hence, it is essential to fabricate multiferroic composites for commercial applications in order to achieve multiferroicity and strong ME coupling. In this study, we have reported the structural characterization, magnetoelectric coupling and magnetic fields dependent dielectric and ac electrical properties of 0.2 $NiFe_2O_4$ (NFO) – 0.8 ZnOnanocomposites prepared by "chemical pyrophoric reaction process" followed by solid state route.

EXPERIMENTAL DETAILS

Multiferroicnanocomposites 0.2 NFO – 0.8 ZnO have been prepared by "chemical pyrophoric reaction process"[4,5] and solid state route. The NFO phase were prepared from high purity $Ni(NO_3)_2.6H_2O$ (Merck) and $Fe(NO_3)_3.9H_2O$ (Merck).The ZnO phase were prepared from high purity $Zn(NO_3)_2.6H_2O$. After

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preparation of NFO and ZnO nanoparticles, we have mixed both the nanoparticles in a requisite amount and grinded it for 10 h. Then the mixed powder has been calcined in air for 5 h at a temperature of 800°C to obtain the required nanometric composite phase. We have made circular pellets from calcinednano powders. Then these pellets have been used for electrical measurements.

Structural characterization has been done through X-ray diffraction technique (XRD) (Bruker D8 Advance Diffractometer) with monochromatic Cu-Kα radiation. Dielectric and ac electrical properties have been carried out using impedance analyzer (KEYSIGHT E4990A). For applied magnetic field, we have used electromagnet (Polytronic Laboratory Electromagnet, EM-100) and electromagnet power supply (Polytronic Power supply, CCP-100Y). We have made an electrode on the sample surface using high temperature silver paste. For ME measurement , we have used homemade ME measurement setup[6].

STRICTURAL CHARACTERIZATION

Structural characterization has been carried out using x-ray diffractometer(XRD) with monochromatic Cu- K_∞ radiation at room temperature. Figure1 shows the XRD pattern of 0.2 NFO– 0.8 ZnOnanocomposites, where the peaks are indexed using JCPDS software having JCPDS PDF No. 441485 and 030891 for NFO and ZnOnanocomposites, respectively. From XRD pattern, we have estimated the average crystallite size (Φ) for both NFO and ZnO phases of the composites using Scherrer formula as $\Phi = k\lambda/\beta\cos\theta$, where, λ is the wavelength of CuK_α radiation (1.542 Å), k is the shape factor, θ is the diffraction angle and β is the full width half maxima of the XRD peak. It is found that the average crystallite size for NFO phase to be ~ 52 nm (±1%) and for ZnO phase ~ 49 nm (±1%).

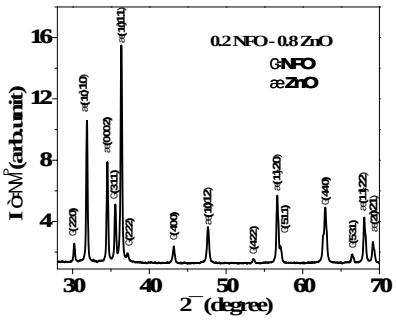

FIG 1: XRD pattern for 0.2 NFO- 0.8 ZnOnanocomposites.

ELECTRICAL PROPERTIES

The magnetic field dependent ac electrical properties of the composite have been investigated using impedance spectroscopy technique. Figure 2(a) shows the variation of Z as a function of frequencies at different magnetic fields. The value of the impedance is found to in the order of kΩ range. The impedance (Z) has been studied at room temperature in presence of applied field (H) of 0, 800 and 2500 Oe. The real part (Z′) and imaginary part(Z″) of impedance are measured on application of H in perpendicular direction of measured electric field. Z vs. *f* curves are found to gradually increase with increase in *f* at different applied H. The observed increased impedance value with increase in H may be due to the magnetostriction property of magnetostrictive materials [7,8]. We have estimated MI for this nanocomposites as shown in inset of Fig. 2(a). The magnetoimpedance (MI) is the change of impedance of a magnetic material under the application of H, which is related as [9] MI (%) = $\frac{Z_H - Z_0}{Z_0} \times 100$ where, Z_H and Z_0 are the impedances under the application of finite H and absence of H, respectively. The observed MI is found to positive, which is ~ 43% at a*f* of 800 Hz. Furthermore, we have plotted magnetic field dependent Nyquist plots as shown in Figs. 2(b). Nyquist plots show two semi-circular arc, one of which lie at high *f* region and the other lie at low *f* region. The observed peak at low *f* region corresponds to the grain boundaries of the sample, whereas the peak at high *f* region corresponds to the grain effect of the sample.

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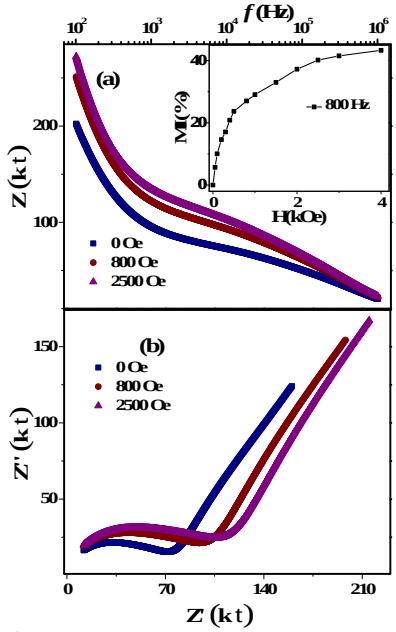

FIG 2: (a) Z vs.*f*,(b) Zð vs. Zå at different magnetic fields, insets in (a) shows MI (%) vs.H plot.

Dielectric Properties

Dielectric constant (ε) as a function of *f* at different H has been measured as shown in Fig. 3(a).εas a function of *f* is found to decrease with increase in H . At high *f* region, ε curves at different H are merged and behave as independent of *f*. High value of ε at low *f* region for different H may be explained through the light of Maxwell Wagner interfacial polarization[10]. Dielectric loss [tan δ] has been studied as a function of *f* at different H as shown in Fig. 3(b). Similarly, tan δ also decreases with increase in H. Magnetodielectric (MD) response has been estimated using the relation as $MD(\%) = \frac{\varepsilon(H,f)-\varepsilon(0,f)}{\varepsilon(0,f)} \times 100$, where, ε(H) is the dielectric constant in presence of H and ε(0) is the dielectric constant in absence of H. However, MD represents a strong coupling between piezoelectric and magnetostrictive phases of the nanocomposites[11]. |MD(%)| at a *f* of 500 Hz is found to ~ 34 % .

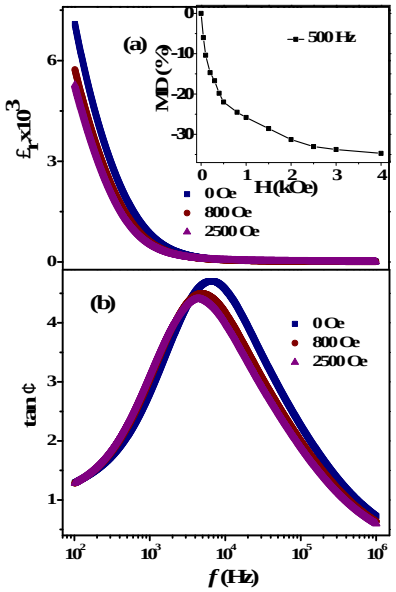

FIG 3: (a) £ vs.*f*,(b) tan ¢ vs. *f* at different magnetic fields, inset in (a) shows MD (%) vs. H curves at *f* = 500 Hz.

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MAGNETOELECTRIC EFFECT

Room temperature magnetoelectric effect (α_E) as a function of H at a*f* of 73 Hz in transverse and longitudinal configurations are shown in Fig. 4(a) and (b), respectively. To obtain ME coupling measurement, the sample has been polled by applying a high voltage across the thickness of the sample. The sample have been placed in a dc H and ac H, where the ac field is superimposed on dc field, the dc H gives a pseudopiezomagnetic effect, which generates an ac electric field (dE = dV/t) across the sample, where, t is the thickness of the sample. α_E has been estimated from an induced voltage dV. The maximum value of α_{E31} and α_{E33} are found to ~3.7 and ~1.04 mV/cmOe at H of 0 Oe and 5 Oe, respectively at a*f* of 73 Hz. α_{E31} vs. H and α_{E33} vs. H show gradual decrease with increase in H. The observed variation of α_E with H for both the configurations may be due to the variation of peizomagnetic coefficient with H of peizomagnetic material of the sample[12]. The decreasing value of α_E with increasing H may be due to the decrease of the growth of the domains with the increasing H leads the decrement of value for both the configurations[13].

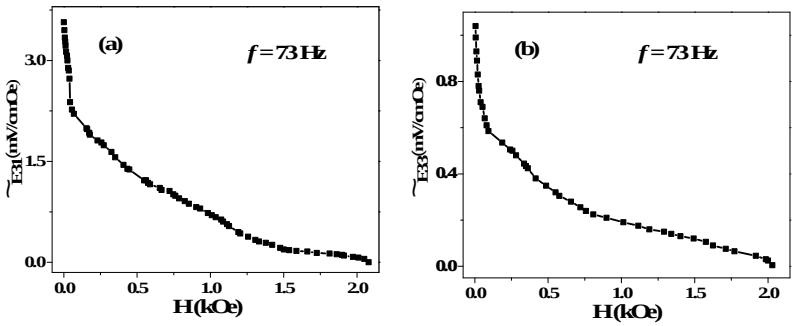

FIG 3: (a) α_{E31} vs.H,(b) α_{E33} vs. H for 0.2 NFO – 0.8 ZnOnanocomposites at a*f* = 73 Hz.

CONCLUSIONS

In the present paper, we have investigated the magnetoelectric coupling and the ac electric transport properties by externally applied magnetic fields. Impedance spectroscopy study shows that the impedance is found to increase with increase in applied H. The calculated MI and MD are found to ~ 43 % at a*f* of 800 Hz and ~34 % at a *f* of 500 Hz. The maximum values of α_{E31} and α_{E33} are found to ~3.7 and ~1.04 mV/cmOe at H of 0 Oe and 5 Oe, respectively at a *f* of 73 Hz.

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