GR-02. Large negative magnetoelectric coupling in Fe substituted ferroelectric $Bi_{0.5}Na_{0.5}TiO_3$ ceramics: A combined experimental and theoretical study. *M. Kumari*[1,2], S. Santapuri[1] and R. Chatterjee[1] *1. Physics Department, IIT Delhi, Hauz Khas, IIT Delhi, New Delhi, India; 2. Department of Applied Physics, Delhi Technological University, New Delhi, India*

In recent years, attention has been paid to synthesize materials with simultaneous spontaneous polarization and a net magnetization, due to their technical applications in spintronics and non-volatile memory [1-2]. Craig J. Fennie proposed a theoretical scheme/criteria to design structures of perovskite oxides ($ATiO_3$, A = Mn, Fe and Ni), such that a polar distortion at Ti-site (B-site of perovskite unit cell) can induce weak ferromagnetism and the direction of magnetization can be switched by application of electric field (known as magnetoelectric (*ME*) coupling) [3]. Following this scheme, in one of our earlier works, we have demonstrated experimentally that a partial substitution of Ti^{4+} (B-site) with Fe^{3+} in a pure ferroelectric system $BaZr_{0.05}Ti_{0.95}O_3$ (BZT) gives rise to a reasonably large *ME* coupling [4] such that the electrical polarization favors the magnetization switching (positive magnetoelctric (*P-ME*) coupling). However, one of the major drawbacks in the BZFT series is that the solid solubility limit of Fe in the main matrix of BZT, is limited only 1.5 % (BZFT15). Thus, in order to have a broader range to test the hypothesis given by Craig J. Fennie, we looked for another strong ferroelectric oxide with no magnetic ordering. BNT ($Bi_{0.5}Na_{0.5}TiO_3$) is reported in literature as an excellent ferroelectric/piezoelectric oxide with large remnant polarization ~38 mC/cm^2 (in comparison of BZT ~21mC/cm^2)[4] and a suitably high Curie temperature 330°C. We substituted Ti with Fe ions systematically in a series of samples, $Bi_{0.5}Na_{0.5}Fe_xTi_{1-3x/4}O_3$ with x = 0.000, 0.005, 0.01, 0.020, 0.050, 0.100 and 0.200. In these samples, solid solubility of Fe substitution could be increased upto 20%. However, an unusual negative ME coupling (*N-ME*) is observed in these samples. To understand this *N-ME* effect, we have developed a Landau-Devonshire thermodynamic model for single phase multiferroic oxides. Thus, in this work, we will present our experimental results for the BNFT series samples along with theoretical modelling to understand the observed *N-ME* effect in these multiferroic oxides.

[1] S.-W. Cheong and M. Mostovoy, Nature Mater. 6, 13 (2007). [2] A. Hill, J. Phys. Chem. B. 104, 6694 (2000). [3] C. J. Fennie, Phys. Rev. B. 100, 167203 (2008). [4] M. Kumari, C. Prakash, R. Chatterjee, Journal of Applied Physics 113, 17D918 (2013).

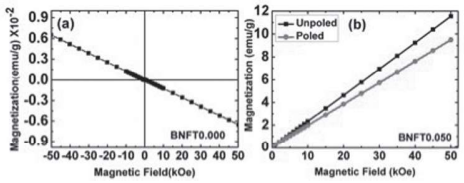


Fig. 1: (a) Room temperature M-H curve for pure BNT and (b) poled and unpoled magnetization virgin curve for x = 0.050 sample.

GR-03. Observation of electric domains in $InMn_{0.75}Fe_{0.25}O_3$: The influence of oxygen vacancies. J. Liu[1,2], *Z. Wang*[1] and S. He[2] *1. Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China, Beijing, China; 2. Department of Physics, Capital Normal University, Beijing 100048, China, Beijing, China*

Recently, hexagonal manganites have attracted increasing attention due to the presence of multiferroicity and topological vortices. Among them, $InMnO_3$ was theoretically expected, and experimentally claimed to show ferroelectricity with ferroelectric transition temperature at about 500 K. Up to date, however, the number of works on its electric domains is still far from adequate. In this work, hexagonal $InMn_{0.75}Fe_{0.25}O_3$ (IMFO) polycrystalline samples were sintered at 1000 °C for 200 hrs by solid state reaction method, and then annealed at 800 °C in following oxygen. It was found that the oxygen annealing slightly increases the weak ferromagnetic components but does not alter the antiferromagnetic long range order temperature at 130 K in the as-sintered IMFO. By contrast, we did clearly observe the domain structures but only in the oxygen annealed IMFO on its polished surface by employing scanning electron microscopy in secondary electron emission mode. Based on the energy-dispersive X-ray spectroscopy mapping and further ferroelectric measurements, we interpret that the electric domain contrasts should not arise from the chemical separation but from the suppressed current leakage due to a reduction of oxygen vacancies in the annealed samples.

GR-04. Phonon Invisibility Driven by Magnetic Ordering in $AlFeO_3$ Thin Film. *S. Tyagi*[1] and V.G. Sathe[1] *1. Raman Laboratory, UGC DAE Consortium for Scientific Research, Indore, India*

AFO crystallizes in two phases, orthorhombic (Pna21) phase with collinear ferrimagnetic structure and Neel temperature (T_N) between 210– 250 K [1] and rhombohedral phase (R-3c) with ferrimagnetic transition T_N at ~225 K [1]. Yosuke et al. [2] described epitaxial growth of metastable multiferroic $AlFeO_3$ film on $SrTiO_3$ (111) substrate which shows enhanced ferrimagnetic transition temperature, T_N = 317K and pinched-like hysteresis loop along with ferroelectricity at room temperature. Thus, AFO can be used as a prospective lead free multiferroic material for the spintronic applications. Therefore, in order to examine coupling of magnetic and lattice degrees of freedom. X-ray diffraction resulted in orthorhombic structure of the grown AFO thin film using pulse laser deposition technique. X-ray reflectivity results gave 30nm thickness and 4.5g/cc material density of the films. Fig. 1 (a) illustrates magnetic measurements as a function of temperature (M vs T) which were carried out in 5-350K temperature range. When cooled, it showed ferrimagnetic behavior up to 280 K as expected for the orthorhombic structure [1]. As the film was cooled, a broad ferrimagnetic to anti-ferromagnetic transition in the 280- 230K temperature range is seen. The anti-ferromagnetic transition is reported only in nano dimensional samples. Fig. 1(b) gives M vs H measurements which further confirms ferrimagnetism at 300K; hysteresis curve showing minor loop and minor saturation, while at 5K, hysteresis curve exhibits composite magnetic behaviour consisting of contributions from soft (ferri) and hard (anti-ferro) magnetic phases. The coexistence of ferrimagnetic and antiferromagnetic orders at low temperatures can be explained by considering grains of different size and shapes in the studied polycrystalline film. The local strain and site disorder enable the ferri- to antiferromagnetic transition in only some particular grains which are of nano dimensions. The temperature dependent Raman spectroscopic study showed phonon re-normalization around the broad magnetic transition which reveals presence of strong spin-lattice coupling in AFO thin film.

[1]. R. Saha et al, Inorg Chem 50, 9527-32, (2011). [2]. Y. Hamasaki et al, Appl. Phys. Lett. 104, 082906, (2014).

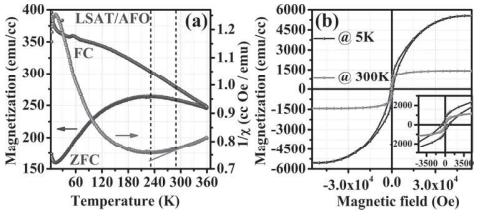


Fig.1. Magnetization of AFO/LSAT film as a function of temperature (a) and applied magnetic field (b).

GR-05. Magnetic field at Ce impurities in La sites of $LaBaMn_2O_6$ double perovskites. *B. Bosch-Santos*[1], N. Nascimento[1], M. Saiki[1], E.L. Correa[1], T.S. Sales[1], L.F. Pereira[1], G.A. Cabrera-Pasca[2] and A.W. Carbonari[1] *1. IPEN/USP, São Paulo, Brazil; 2. Física, Universidade Federal do Pará-Campus Abaetetuba, Abaetetuba, Brazil*

Magnetic behavior in LaBaMn2O6 double perovskite compounds has been investigated with various techniques, due to the rich variety of electromagnetic properties, such as a colossal magnetoresistance, charge and orbital

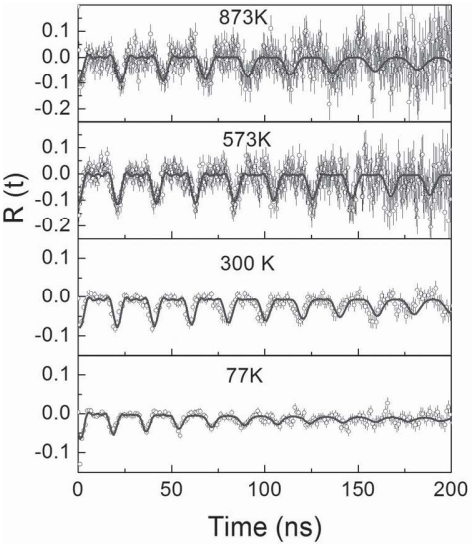
ordering, and metal-insulator transition. In this paper, we have used a nuclear and short-range technique, the Perturbed Angular Correlation (PAC) spectroscopy, to investigate the magnetic hyperfine field at the 140Ce nucleus of Ce impurities occupying La sites. The radioactive 140La nuclei with a half-life of 40.8 h were produced by direct activation of natural La present in the samples through the irradiation with neutrons in the IEA-R1 nuclear research reactor of IPEN. The PAC measurements were carried out with a six BaF2 detector spectrometer at several temperatures between 10 K and 400 K. This double perovskite samples were synthesized by sol-gel route. The crystal structure was determined by X-ray diffraction and the analyses showed that this method produced perovskite oxides with cubic structure in Pm-3m space group. This phase occurs due to an oxygen deficiency. The local properties investigated by PAC spectroscopy revealed a ferromagnetic transition temperature above 300 K and an anomalous behavior of the temperature dependence of magnetic hyperfine field at La sites, which can be ascribed to the contribution of 4f band of Ce to Bhf at low temperatures due to the increase in its localized character.

GR-06. Engineering Magnetoresistance in (001)- and (111)-oriented $(La_{2/3}Sr_{1/3}MnO_3)_n/(LaFeO_3)_{10}$ Superlattices through Interfacial Coupling. Y. Zhou[1,2] and *S.J. May*[2] *1. Materials Science and Engineering, Shandong University, Ji'nan, China; 2. Materials Science and Engineering, Drexel University, Philadelphia, PA, United States*

In ABO_3-based heterostructures, the coupling of BO_6 octahedral rotations across interfaces has been shown to play a significant role in magnetic ordering temperatures and magnetocrystalline anisotropy in complex oxide films. In this work, we synthesize and characterize $(La_{2/3}Sr_{1/3}MnO_3)_n/(LaFeO_3)_{10}$ superlattices, in which we vary the crystallographic orientation and the thickness of thin $La_{2/3}Sr_{1/3}MnO_3$ (LSMO) layers in order to investigate the role of octahedral coupling on magnetoresistance in manganites. The octahedral rotations in bulk $LaFeO_3$ are larger than those in bulk LSMO, which we hypothesize results in suppressed on the Mn-O-Mn bond angles in the interfacial regions of LSMO due to interfacial octahedral coupling. Oxide molecular beam epitaxy was used to deposit superlattices with $n = 7$, 10, and 14 on both (001)- and (111)-oriented $SrTiO_3$. The different crystallographic orientations allow for the octahedral coupling at the interface to be tuned, with weaker coupling anticipated for the (001)-interface than the (111)-interface as the *B*-site cations are bridged by 1 and 3 oxygen atoms, respectively, at these interfaces. In the $n = 14$ superlattices, the effect of orientation is minimal with both (001)- and (111)-oriented samples exhibiting similar Curie temperatures and magnetoresistance. As the ratio of interfacial to non-interfacial volume within the LSMO layers is increased by decreasing *n*, we find that the transition temperatures and the magnetoresistive behavior of the samples diverges. Larger ordering temperatures and reduced magnetoresistance are found in the (001) superlattices compared to the (111) superlattices. This result is consistent with octahedral coupling playing a more significant role at (111)-heterointerfaces, and points to a structure-driven approach to tuning interfacial magnetoresistance in complex oxide heterostructures. Yu Zhou was sponsored by the China Scholarship Council (CSC).

GR-07. Study of the Local Magnetics and Electrics Properties at $Gd_2Ti_2O_7$ by PAC Spectroscopy and ab Initio Calculations. E.L. Correa[1], L.F. Pereira[1], *W.L. Ferreira*[1], B. Bosch-Santos[1], L. Scalise[1], V. Gonçalves[1], R. dos Santos[1], J. Schell[2] and A.W. Carbonari[1,2] *1. Instituto de Pesquisas Energéticas e Nucleares, Universidade de São Paulo, São Paulo, Brazil; 2. ISOLDE, CERN, Geneve, Switzerland*

$Gd_2Ti_2O_7$ exhibits a complex magnetic behavior with a geometrically frustrated antiferromagnetic ordering below $T_N = 1.1$ K and a second magnetic transition at 0.7 K. In the work here reported, perturbed angular correlation (PAC) technique was used in order to measure electric quadrupole hyperfine interactions at ^{111m}Cd probe nuclei on Ti sites. These experimental results along with those for magnetic hyperfine interactions at 155Gd previously reported from Mossbauer Effect spectroscopy were interpreted with electronic structure first-principles calculation. Single and doped-Cd cell simulations have been performed with potential linearized augmented plane waves methods and generalized gradient approximation from Perdew-Burke-Ernzerhorf embodied within the WIEN2k all-electron code. PAC measurements have exhibited ^{111m}Cd probes occupying two fraction sites characterized by well-defined quadrupole frequencies (as shown in the spin-rotation spectra in the figure) with almost the same values for the corresponding electric field gradient (efg), however, with slightly difference in the asymmetry parameters. Theoretical efg's obtained for Cadmium atoms located at Gd or Ti sites of the pyrochlore structure indicate that both experimental hyperfine parameters corresponding to Cd at Titanium site.



GR-08. Tunneling Magneto-Dielectric Effects of Crystalized Co-BaF_2 Nano-granular Films at MHz Frequencies. *H. Kijima-Aoki*[1], Y. Cao[1], Y. Endo[1], N. Kobayashi[2], S. Ohnuma[2,1] and H. Masumoto[1] *1. Tohoku University, Sendai, Japan; 2. Research Institute for Electromagnetic Materials, Tomiya, Japan*

Magneto-dielectric effect is of great interest to application for electromagnetic device due to their low dissipation power and high frequency response. We have reported that the permittivity (ε) change by an external magnetic field (*H*), namely tunneling magneto-dielectric (TMD) effect, observed at room temperature in metal-insulator type nano-granular film[1]. Fluoride-based nano-granular films has potential to achieve high TMD effect due to their high electrical resistivity (ρ) and low tanδ(δ=ε″/ε′). Since an intergranular matrix by the metal-fluoride such as MgF_2[1] and AlF_2[2] has high stability of crystallization (high heat of formation energy), the film easily forms nano-granular structure. The origin of TMD effect has been accounted by spin-dependent charge oscillation caused by quantum mechanical tunneling between magnetic granules[3], so inter-granular matrix structure and its electrical properties are important. In this report, we have focused on crystal BaF_2 as intergranular matrix with dielectric constant of 7.8 easy to form ionic crystals, and structure and TMD effects of Co- BaF_2 nano-granular system film with highly crystallized BaF_2matrix have investigated. A series of Co-BaF_2 films with Co content from 0 to 42 at.% were prepared by rf-magnetron sputtering, where Co granules of 2-3 nm in diameter are dispersed in BaF_2 matrix. Intergranular BaF_2 matrix is well crystallized and its (1 1 1) axis is preferentially oriented to out of plane. From *M-H* curves, Co-BaF_2 films show superparamagnetic behavior. The ρ of Co-BaF_2 films with 42 at.%Co film is still high about 30 Ω2cm enough to measure ε. TMD ratio ($\Delta\varepsilon/\varepsilon_0$) was evaluated $(\varepsilon_{10kOe}-\varepsilon_0)/\varepsilon_0$, where ε_0 and ε_{10kOe} is ε under zero-field and a in-plane magnetic field of 10kOe, respectively. The frequency where the maximum TMD effect increases from 400k to 1MHz with increasing Co contents. The maximum $\Delta\varepsilon/\varepsilon_0$ of Co-BaF_2 films is found to be 2-4 at.% for 26-42 at.%Co. The magnetic field dependence of $\Delta\varepsilon/\varepsilon_0$ is reasonably fitted to $(M/M_{10k})^2$-*H* curves, supported by spin-dependent tunneling process in nano-granular film[3].