Strain induced giant magnetoelectric coupling in KNN/Metglas/KNN sandwich multilayers

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A lead-free magnetoelectric composite with sandwich layers of $K_{0.5}Na_{0.5}NbO_3$ (KNN)/$Co_{76}Fe_{14}Ni_4Si_5B$ (Metglas)/KNN is fabricated as a cantilever and it is characterized for its magnetic, ferroelectric, and magnetoelectric properties. Giant magnetoelectric (ME) coupling is recorded under both resonant and sub resonant conditions and the data are presented here. The observed magnetoelectric coupling coefficient reaches a maximum of 1321 V/cm Oe at resonance (750 Hz) and 9.5 V/cm Oe at a sub-resonant frequency of 50 Hz. The corresponding theoretical calculations are provided for comparison. High magnetostriction as well as initial permeability, fairly good piezoelectric properties, and low dielectric constant cumulatively contribute to the giant magnetoelectric properties in the present system. The high resonance and sub resonance ME coupling voltages make the system ideal for transducers and energy harvesting device applications. *Published by AIP Publishing.* [http://dx.doi.org/10.1063/1.4973450]

The field of research attracts considerable scientific attention on magnetoelectric (ME) and multiferroic materials because of their enticing aspect of developing new multistate memory devices, magnetoelectric sensors, transducers, and multimodal energy harvesters.[1,2] The combined effect of both ferroelectric and ferromagnetic orders in this type of material can accelerate the progress of multifunctional devices which are expected to replace the conventional electromagnetic devices. Magnetoelectric coupling effect, the interesting aspect of multiferroics, is strongly observed in composite materials than in single phase multiferroic materials especially at room temperature.[3] Strain/stress mediated ME coupling is offered by the magnetoelectric composites having distinct ferromagnetic and ferroelectric/piezoelectric components combined in different geometries.[4] These materials are originally proposed for use in magnetic field sensors, but now the renaissance of research in ME composite heterostructures also proved their technological importance in high-voltage harvesting devices in wireless electronics.

The ease of synthesis techniques and their easy integration into devices make 2-2 multilayer thin films more preferred to achieve better magnetoelectric coupling responses.[5] Also, these layered composite structures are reported to be less sensitive to leakage current with high magnetoelectric coupling properties.[6] But the substrate clamping effect can reduce the ME coupling to several orders.[7] Giant magnetoelectric coupling in composite materials is mostly reported for Lead Zirconate Titanate (PZT)/alloy composites such as PZT/Metglas[8] and PZT/Terfenol-D[9] heterostructures than PZT/ferrites.[10] Unlike ferrites, the magnetic properties of alloys can be effectively tuned by tuning their composition. Soft magnetic alloys like Metglas, permalloys, and Terfenol-D are widely used nowadays in ME composites due to their extremely high permeability and enhanced magnetostriction.[11]

Here, authors fabricated a tri-layered lead-free magnetoelectric 2-2 composite system where Metglas thin films are sandwiched between two layers of piezoelectric $K_{0.5}Na_{0.5}NbO_3$ (KNN) thin films. KNN is the most sought after lead-free piezoelectric candidate with properties comparable to PZT. High Curie temperature (T_c), and fair piezoelectric and ferroelectric properties make KNN bulk and thin films interesting.[12–15] For getting giant magnetoelectric voltages, the magnetic material in the composites should be highly magnetostrictive. Metglas is one among the best candidates which possesses high initial permeability (>400 000) and magnetostriction (20–25 ppm)[16,17] which is already proved to be a good option as a magnetic component in ME composites.[18]

In the present investigation, top and bottom layers of the sandwich structures are fabricated by two methods, sputtering and sol-gel spin coating. The base KNN layer is deposited using the RF sputtering technique. The KNN target for sputtering is prepared via the ceramic method by employing metal carbonates of sodium and potassium as well as oxide of Niobium. The powders are pre-sintered at 400 °C for 1 h. The as-prepared KNN powders are pressed into pellets of 2 in. diameter under a pressure of 8 tons and are sintered at 700 °C for 6 h which is used as the target for sputtering KNN. KNN thin films are sputtered on an ultrasonically cleaned surface of the $Si/SiO_2/Ti/Pt$ substrate. The sputtered KNN thin films are pyrolyzed at a temperature 200 °C for 30 min and are annealed at 700 °C for 1 h in vacuum. The top most layer of KNN on the magnetic layer is fabricated by a sol-gel based spin coating method (6000 rpm for 30 s) employing potassium, sodium, and niobium ethoxide as raw materials and 2-methoxy ethanol as the solvent. The middle magnetic (Metglas) layer is deposited on the substrate by RF sputtering using commercially available Metglas ribbons

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