1-1-2012

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Recommended Citation
Journal of Applied Physics 111, 7C720 (2012)

Journal of Applied Physics

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Citation: J. Appl. Phys. **111**, 07C720 (2012); doi: 10.1063/1.3679042
View online: http://dx.doi.org/10.1063/1.3679042
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v111/i7
Published by the American Institute of Physics.

JOURNAL OF APPLIED PHYSICS **111**, 07C720 (2012)

Magnetoelectric effect in AlN/CoFe bi-layer thin film composites

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(Presented 3 November 2011; received 23 September 2011; accepted 1 December 2011; published online 9 March 2012)

The present work is aimed at fabricating bi-layer aluminum nitride (AlN)/cobalt iron (CoFe) magnetoelectric (ME) thin films using reactive rf/dc magnetron sputtering. A systematic study on structural, morphological, piezoelectric, magnetic and magnetoelectric properties is undertaken. Except for AlN and CoFe, no other phases were detected with the layer thicknesses measured at 160 and 130 nm, respectively. The rms roughness measured was around 2.096 nm for AlN and 1.806 nm for CoFe. The bi-layer thin film exhibited both good piezoelectricity and ferromagnetism, as well as ME effect. A 52% change observed in the piezoelectric signal, measured using magnetic field assisted piezoresponse force microscopy, can be ascribed to the existence of a stress-mediated magnetoelectric coupling between AlN and CoFe. © *2012 American Institute of Physics*. [doi:10.1063/1.3679042]

I. INTRODUCTION

Magnetoelectric (ME) effect, the induction of magnetization by an electric field or polarization by a magnetic field, attracted the vision of researchers worldwide owing to the coupling between electric and magnetic order parameters and the potential to manipulate one through the other in a number of high technology applications.[1,2] New "stress mediated" composite materials mainly comprising magnetostrictive and piezoelectric or electrostrictive based materials displayed ME effects several orders of magnitude higher than single phase ME materials. Most of these synthesized composites exist in the form of bulk materials, and therefore are hard to be integrated into micro-electro-mechanical-systems (MEMS) or microelectronic devices.[3,4] Nevertheless, recent investigations on ME thin film multilayers and epitaxial heterostructures grown on single crystal substrates displayed very promising features.[5] Aluminum nitride (AlN) is a lead-free piezoelectric material with wurtzite structure. It is a good candidate for use in high frequency applications owing to its low leakage current, low dielectric constant, and low dielectric losses in microwave and millimeter wave frequencies.[6,7] It has been widely investigated for its usage in microelectronic and optoelectronic devices such as thin film bulk acoustic wave resonators (TFBARs), short wavelength emitters, and electronic packaging. Piezoresponse force microscopy (PFM) has long been used to image domain structure in the ferroelectric thin films and polarity distribution in the III-nitride films with nanometer scale spatial resolution.[8–10] Cobalt iron ($Co_{50}Fe_{50}$) is a soft magnetic material used in technical applications that have high permeability, high saturation magnetization, high positive magnetostriction coefficient, and high Curie temperature.[11] Even though these two materials have been investigated independently, combined effects of their heterostructures are unknown. A thorough literature search on the properties of AlN/CoFe bi-layers did not yield any positive results. As a key product property of such a lead-free bi-layer structure, magnetoelectric effect (ME) has a great potential in sensors and other applications. Therefore, it is worth studying the important aspects of piezoelectric-magnetic interactions in the bi-layers of piezoelectric AlN and magnetostrictive CoFe. In the present investigation, we use magnetic-field-dependent PFM as a direct probe to stress mediated magnetoelectric coupling in AlN/CoFe thin films. Our study reveals marked magnetic field-induced changes in the piezoelectric amplitude signal of the bi-layer AlN/CoFe film, which may be caused by the mechanically transferred strain from the magnetostrictive CoFe layer.

II. EXPERIMENTAL

Magnetoelectric aluminum nitride/cobalt iron, AlN/CoFe, bi-layer thin films were fabricated using reactive dc/rf magnetron sputtering (ATC 1300 dc and rf magnetron sputtering system, AJA International, MA). First, aluminum nitride films were deposited at 300 °C on a Si (111) wafer by rf magnetron sputtering using an aluminum target (99.999%) using pure nitrogen (N_2) and argon (Ar) in an ultrahigh vacuum chamber at a background pressure $\geq 3.6 \times 10^{-8}$ Torr. The working pressure during the deposition was 6 mTorr with 18 SCCM (or cubic centimeter per minute at STP) flow of ultrahigh pure argon and nitrogen gases in 1:1 ratio. Subsequently, cobalt iron films were grown on the top of aluminum nitride layer by dc magnetron sputtering at 100 °C. A thin 10 nm copper layer was deposited as a capping layer to prevent the oxidation of CoFe. The working pressure during the deposition of CoFe layer was 3 mTorr with 18 SCCM flow of Ar gas. The deposition rates were: 0.88 nm/min and 4.3 nm/min for an AlN and CoFe layers at 250 W (rf power) and 100 W (dc power), respectively.

The phase purity and the crystal structure of the films were analyzed by x-ray diffraction using a Rigaku MiniFlex

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II x-ray diffractometer with Cu kα radiation. The surface morphology and the microstructure of the films were investigated with a LEO 1530 VP field emission (FE) scanning electron microscope (SEM). Topology images of the AlN films were collected at room temperature with an Asylum Research MFP-3 D atomic force microscope working in tapping mode and using commercial Si_3N_4 cantilevers with a force constant of 9.7 N m^{-1}. The piezoelectric signal of the samples was measured with the high voltage PFM module of the MFP-3D AFM by using an AC240TM cantilever made of a tetrahedral silicon tip coated with Pt/Ti. The magnetoelectric measurements (magnetic field assisted PFM) carried out using the same system but additionally applying an in-plane magnetic field with a variable field module. Magnetic nature of the films was examined with Princeton Measurement Corporation MicroMag^{TM} 3900 vibrating sample magnetometer at room temperature.

III. RESULTS AND DISCUSSION

Figure 1 shows the x-ray diffraction patterns of the AlN/CoFe bi-layer thin film fabricated on Si (111) wafer. All the peaks can be indexed to parent phases. No peaks other than the AlN and CoFe phase are observed in the figure. Figure 1 also indicates polycrystalline nature of both AlN and CoFe present in the bi-layers. The surface morphology and thicknesses of the films were examined by FE-SEM. These SEM studies indicate that the deposited films are smooth, crack-free, and with a good homogeneity. The thickness of AlN and CoFe layers found to be 160 and 130 nm, respectively. Insets of Figs. 1(a) and 1(b) show the AFM topology and PFM images of AlN thin film. AFM images indicate that the film is uniform with densely packed spherical grains of about 200 nm. The rms roughness of the thin film is 2.906 nm. The piezoresponse force microscopy (PFM) contrast image of AlN film reveals a random distribution of bright and dark regions all across the film surface indicating the polarity distribution present in the film.[10]

Magnetization measurements carried out both in-plane of out of plane directions at room temperature for CoFe and AlN/CoFe bi-layer thin films. Figure 2 displays the normalized magnetization curves for CoFe and AlN/CoFe bi-layer. We can see the coercivity is larger for AlN/CoFe bi-layer (45 Oe) compared to CoFe (23 Oe). This may have been caused by the interfacial anisotropy as a result of lattice mismatches between Si and CoFe and AlN and CoFe.[12]

AlN grows in the wurtzite structure with the polar direction along the *c*-axis. As a polar material, an AlN film with the *c*-axis perpendicular to the substrate can have two physical orientations of the columnar grains with the film surface terminating either in the aluminum or nitrogen basal plane. A poly crystalline film, therefore, can have a mixture of these two orientations.[13] It is well known that, in the wurtzite III-nitrides the spontaneous polarization points from N atom to the nearest neighbor III-metal atom(anion to cation) along the crystallographic *c*-axis, therefore, we believe the upward (N polarity) and downward (Al polarity) spontaneous polarizations were distinctively represented by dark (out of phase) and bright (in phase) areas, respectively.[10,14] In Fig. 1(b), the bright areas correspond to grains with the normal component of the polarization pointing toward the film surface (Al polarity), whereas the dark spots indicate areas where the polarization is oriented in the opposite direction (N polarity). In contrast to ferroelectric materials, the orientation of the spontaneous polarization in nitrides cannot be changed by the application of an external electric field. While the predicted spontaneous polarization is independent of strain, piezoelectric polarization is strain-induced.[14] In general, the total polarization of a nitride layer is the sum of the spontaneous and of the piezoelectric polarizations.[15]

The piezoelectric response dependence of applied voltage for 160 nm thick AlN thin film is plotted in Fig. 3. Both the phase and amplitude curves confirm the existence of polarization in AlN layer at room temperature. The observed piezoelectric response of the AlN film is the result of the polarity distribution. The film exhibit a well-known butterfly type behavior seen in strain versus field curves, for the variation of amplitude versus the voltage with a maximum displacement of 0.31 nm at a driving voltage of V = + 42.24 V. The effective value of zero field piezoelectric coefficients (d_{33}) of the AlN film is calculated by fitting the liner portion of the butterfly loop. It is well known that the amplitude of the displacement of the tip (A) is proportional to the ac bias

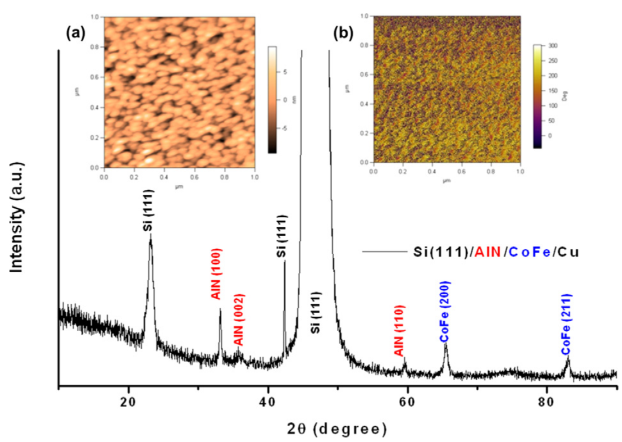

FIG. 1. (Color online) XRD patterns of AlN/CoFe bi-layer ME composite thin film. Inset shows (a) topography AFM image, (b) phase contrast image of 130 nm AlN thin film.

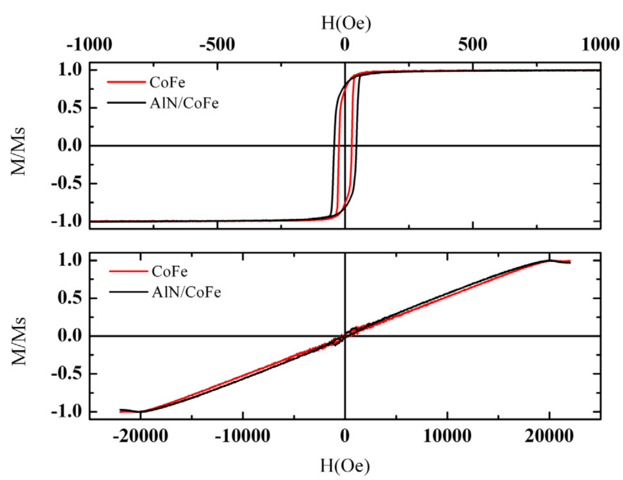

FIG. 2. (Color online) Magnetic hysteresis loops of CoFe and AlN/CoFe bi-layer.

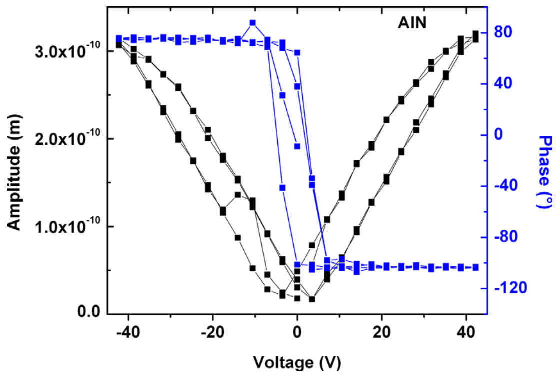

FIG. 3. (Color online) The piezoelectric response dependence of applied voltage for the AlN thin film fabricated at 300 °C.

voltage (V_{ac}): A = $d_{33}V_{ac}Q$,[16] where Q is a proportionality constant that varies from 10 to 100 and accounts for the amplitude enhancement at the tip-sample resonance. By considering the value of the proportionality factor at the lower end of this range, the magnitude d_{33} calculated for the AlN is 0.74 pm/V. The value is in good agreement with literature reports for polycrystalline AlN thin films.[17]

The variation of the amplitude of the local piezoelectric response of AlN/CoFe bi-layer films under different bias magnetic fields, for driving voltages of V = ±22.4 and ± 42.24 V, are presented in Fig. 4. In general, the PFM hysteresis loops are asymmetric with respect to the origin, presumably due to defects, the surface charges at the electrode and film, the different barrier potential between the electrode and the film, the presence of an internal electric field inside the film or pinning of the domain walls by defects.[18–20] In the present case, the asymmetric behavior in piezoresponse of AlN/CoFe at 0 Oe, Fig. 4, is likely due to the surface charges at the electrode and CoFe as reported in literature.[20] When the bias magnetic field (*H*) is applied in parallel to in-plane of AlN/CoFe bi-layer, due to magnetostriction the magnetic domains reorient in the CoFe film. The CoFe possesses large positive linear magnetostriction. Hence, under the applied magnetic field CoFe would expand a bit in *H* direction, but shrink a bit in the direction

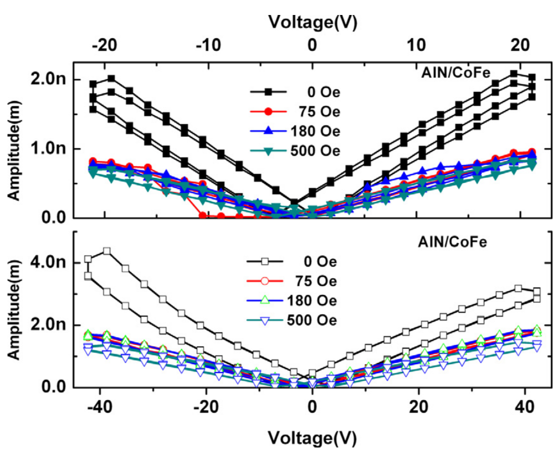

FIG. 4. (Color online) The piezoelectric response amplitude variations with applied bias magnetic field, with driving voltage of V = ±22.4 and ±42.24 V, for AlN/CoFe bi-layer.

normal to the *H*. The magnetostriction of the top layer of CoFe would impose a stress on the bottom AlN film. The stress mediated ME coupling between AlN and CoFe layers evidenced from the change in the piezoelectric amplitude signal under bias magnetic field. The amplitude of the piezoelectric signal decreases from 3.09 to 1.42 nm, which corresponds to the 52% change of the signal (in amplitude versus voltage measurements@ V=+42.24 V) caused by the increasing the bias magnetic field from 0 to 500 Oe. This indicates the strong ME coupling between piezoelectric AlN and magnetostrictive CoFe layers. Similar trend is also reported in $PbTiO_3$-TbDyFe bi-layer nano composites.[21]

IV. CONCLUSIONS

In summary, lead-free magnetoelectric bi-layer thin film of AlN/CoFe was fabricated successfully with rf/dc magnetron sputtering. The bi-layer possesses both electrical and magnetic properties at room temperature reflecting the properties of their parent compounds. Strong stress mediated magnetoelectric coupling in the bi-layer is evidenced from 52% change in the piezoelectric signal in magnetic field assisted piezoresponse force microscopy measurements.

ACKNOWLEDGMENTS

This work was supported by the Defense Advanced Research Projects Agency (DARPA) under Grant No. HR 0011-09-1-0047.

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