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Deposition of magnetoelectric hexaferrite thin films on substrates of silicon

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ABSTRACT

Magnetoelectric M-type hexaferrite thin films ($SrCo_2Ti_2Fe_8O_{19}$) were deposited using Pulsed Laser Deposition (PLD) technique on Silicon substrate. A conductive oxide layer of Indium-Tin Oxide (ITO) was deposited as a buffer layer with the dual purposes of 1) to reduce lattice mismatch between the film and silicon and 2) to lower applied voltages to observe magnetoelectric effects at room temperature on Silicon based devices. The film exhibited magnetoelectric effects as confirmed by vibrating sample magnetometer (VSM) techniques in voltages as low as 0.5 V. Without the oxide conductive layer the required voltages to observe magnetoelectric effects was typically about 1000 times larger. The magnetoelectric thin films were characterized by X-ray diffractometer, scanning electron microscope, energy-dispersive spectroscopy, vibrating sample magnetometer, and ferromagnetic resonance techniques. We measured saturation magnetization of 650 G, and coercive field of about 150 Oe for these thin films. The change in remanence magnetization was measured in the presence of DC voltages and the changes in remanence were in the order of 15% with the application of only 0.5 V (DC voltage). We deduced a magnetoelectric coupling, α, of 1.36×10^{-9} s m^{-1} in $SrCo_2Ti_2Fe_8O_{19}$ thin films.

1. Introduction

Single-phase magnetoelectric (ME) hexaferrites have exhibited high ME coupling, α, at room temperature comparable or better than composites of ME multi-layers [1–12]. The ME effect implies that the application of a magnetic field, *H*, induces an electric Polarization, *P*, and in the converse case applying an electric field, *E*, magnetization, *M*, in the material is induced. In most cases, substitution of Sr ion for Ba ion in Z-type hexaferrites (this applies to other hexaferrites as well) strains the bonding of the chemical combination of Fe–O–Fe located near Sr ion, resulting in magnetic moments canted with respect to the c axis of the hexaferrite and formation of a spin-spiral [13]. The application of *E* strains the material and thereby changes the physical structure of the spiral spin configuration. It is this physical motion of the spiral response to *E* that induces a change in magnetization, *M*. In previous work most of the research has been concerned with the electric and magnetic properties of bulk properties of ME hexaferrite materials [1–10]. However, recently M-type hexaferrite films have been deposited successfully in the Microwave laboratory [14] at Northeastern University by pulsed laser deposition, PLD, technique. The deposited films exhibited ME effects at room temperature with ME coupling, $\alpha = 6.07 \times 10^{-9}$ s/m measured by a vibrating sample magnetometer (VSM) technique by applying the voltage to the film ranged from 500 to 1000 V. Voltages of that amplitude are simply not compatible with planar technologies of IC circuits on CMOS devices.

Recently, new techniques for *H*-field and *E*-field sensing as well as tunability applications in RF devices were explored [15,16] using single-phase hexaferrite films of $SrCo_2Ti_2Fe_8O_{19}$, on sapphire substrate. To make these new kind of devices compatible with IC technology, the applied voltage required was reduced by applying *E* fields in the film plane in a multi capacitive structure [15–17] whereby parallel metallic strips were deposited on the ME film. On the other hands, in order to reduce the required voltage for the case that *E* field applied perpendicular to the film, M-type hexaferrite films were successfully deposited on a conductive oxide layer, Indium Tin Oxide (ITO) which also acts as buffer layer between the ME film and sapphire substrate. In order to incorporate these films into the new generation of ME devices and integrate them with CMOS technology two main obstacles must be resolved: 1) there was a need to reduce the required voltage to generate practical E fields for inducing measurable ME effects at room temperature. 2) A need to "make" ME films compatible to Silicon wafers. Thus, we have embarked on a search for buffer layer materials that accomplished the two stated above goals.

ITO buffer layers were utilized to deposit the M-type hexaferrite films, $SrCo_2Ti_2Fe_8O_{19}$, on silicon. We expected that the lattice constant mismatch between a metallic buffer layer and a

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ME film was too great to overcome in depositing ME films. However, conductive oxide buffer layers of ITO (In_2O_3: SnO_2) and AZO (Al_2O_3: ZnO) have a better match to the lattice constant of the ME film (5.9 Å for hexaferrites, 5.25 Å for ZnO, and 10.12 Å for ITO, about twice that of hexaferrites). The introduction of metallic (Au and Cu) and AZO as buffer layers was not successful in depositing ME films on silicon wafers. However, we have carried out successful deposition of ME films on silicon by introducing the ITO conductive oxide buffer layer which was essential in depositing ME hexaferrite films. In our previous work, we successfully deposited the ME film on ITO with sapphire substrate [18]. This deposition feasibility allows for the possibility of both reducing required voltages to observe ME effects in devices and make them compatible with CMOS devices on silicon for applications, such as sensors, filters, IC chips, magnetic recording control valves and tunable inductors. These types of devices require that the ME films to be part of the integrated circuitry.

2. Thin film deposition

Targets of $SrCo_2Ti_2Fe_8O_{19}$ were prepared by conventional ceramics processing [14,19]. The chemical compositions of the target were verified by X-ray diffraction (XRD), using a CuKa source, and energy dispersive X-ray spectroscopy (EDXS) and it was determined to be single-phase material. Indium Tin Oxide, In_2O_3/SnO_2 (ITO) target was purchased from Sigma Aldrich. Silicon wafers with orientation of $<111>$ was oxidized in a wet oxidation oven with a thickness of 500 nm for isolation purpose. Both ITO and $SrCo_2Ti_2Fe_8O_{19}$ films were deposited on Si/SiO_2 substrate by PLD technique, which is an effective epitaxial growth technique for the production of hexaferrite thin films [20].

The base pressure in the deposition chamber was maintained at 9×10^{-6} Torr and for ITO deposition the substrates were heated to 400 °C. The films were deposited in a high purity oxygen environment of 10 mTorr. A KrF excimer laser with a wavelength of 248 nm and energy of 400 mJ/pulse was focused on the target surface. The distance between the target and the substrate was set to 5 cm and the repetition rate of the laser was set to 10 Hz during deposition and it lasted for 20 min. After deposition, the films were cooled to room temperature at the same oxygen pressure. The vacuum was broken to mask part of the sample in order to have access to ITO later after the ME film deposition. The substrate was heated to 600 °C during the deposition of the ME film in a high purity oxygen environment of 200 mTorr. The repetition rate of the laser gradually increased from 1 Hz to 10 Hz during deposition to improve the growth. The deposition run was timed for 60 min and resulted in an amorphous film structure as confirmed by XRD. The film thickness was measured to be about 1 μm using a scanning surface profilometer. The films were inserted into a preheated furnace of 1050 °C, annealed for 40 min, and rapidly removed from the furnace. Leaving the films in the furnace during temperature ramping appeared to affect crystal structure due to diffusion at the film-substrate interface. The thin films were evaluated by XRD and EDXS to determine the crystal structure and composition of the film and buffer layer.

3. Experimental results

3.1. Magnetic characterization of the films

The composition and structure of these films were determined by X-ray diffractometer, energy dispersive X-ray spectroscopy, and SEM image. Fig. 1a is the SEM data of a film tilted 30°. Fig. 1b is the magnified image of the same part in Fig. 1a. The hexagonal polycrystalline structures are visible in images that show each grain is made of hexagonal unit cells of the ferrite material. The XRD patterns are illustrated in Fig. 2. It shows that the film exhibited a polycrystalline structure but has strong and sharp diffraction peaks at (0 0 2 n) [*n* is 3, 4, …] as shown in Fig. 2, other peaks correspond to other crystal phases, such as, amorphous thermally grown SiO_2, Si and the buffer conductive layer of ITO deposited in between the SiO_2 layer and the ME film. For example, the peak at 69° corresponds to Si and the peak at 32° has been observed on any film and any substrate (Si/SO_2 or Sapphire) but with the ITO buffer layer. However, in addition to these aforementioned background phases the interfacial region between ME film and ITO layer may consist of extraneous magnetic phase, not consistent with the M-type crystal structure. Since the film was deposited at relatively low temperatures (600 °C), we estimate this interfacial region to be in the order of 50–80 nm [21] which is much smaller than the ME film thickness of 1000–1300 nm.

The static magnetic properties of the films were studied by vibrating sample magnetometer (VSM) technique with the magnetic field applied perpendicular and parallel to the film plane. In Fig. 3, typical hysteresis loops of the films are shown. The coercive field for the two cases that the external field applied parallel or perpendicular to the film plane was measured to be 214 Oe and 130 Oe, respectively. We measured saturation magnetization of 650 G.

3.2. Magnetoelectric measurements

We performed ME measurements by measuring the changes in remanence magnetization with the application of a DC voltage, see Fig. 4. The ME effect at room temperature was observed at very small DC voltages in films of $SrCo_2Ti_2Fe_8O_{19}$/ITO as the voltage is being directly applied to the thickness of the film, about a micron, provided by conductive ITO layer, and top silver conductive layer.

In Fig. 5, the magnetization (magnetic flux generated within the film) is plotted as a function of applied voltage across the film.

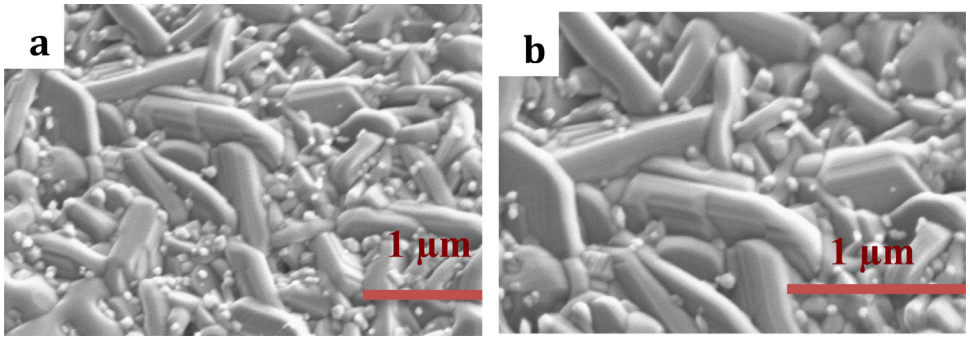

Fig. 1. SEM image of the $SrCo_2Ti_2Fe_8O_{19}$ thin film on Si/SiO_2/ITO substrate.