Co Thickness Effect on the Dielectric Permittivity of SiO_2/Co/SiO_2 Films

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The effect of Co inserted layer with thickness below 20 nm on the dielectric permittivity of SiO_2(60 nm)/Co(x nm)/SiO_2(60 nm) thin films fabricated on glass B270 substrates by the reactive sputtering technique was studied. The dielectric constant is around 7.4 for B270 glass substrate and SiO_2/B270 film. However, it is rapidly raised up to roughly 55 for all the SiO_2/Co/SiO_2 samples with the thickness of Co inserted layer larger than 2 nm. From the cross section TEM pictures of the SiO_2/Co/SiO_2 films with 1 and 2 nm Co thickness, we have experimentally demonstrated that this enhancement behavior of the dielectric constant is due to the growth mechanism of the Cobalt inter-layer from island clusters to continuous Co layer for samples with x larger than 2 nm. The adding of a Co inter-layer redistributes the interface charges between Co and SiO_2 layers, and that enhances both the intrinsic polarization and its dielectric constant. For the magnetic induced dielectric variation, the variation of the dielectric constant also increased with thickness of Co for samples with x larger than 2 nm. A direct observation of a 0.04–0.20% dielectric variation is induced by external magnetic field. However this increase behavior is roughly saturated for applied magnetic field roughly above 60 Oe. The magnetodielectric properties in SiO_2/Co/SiO_2 films are manifested and it has potential for a ferroic sensor application.

***Index Terms*—Cobalt (Co), dielectric constant, magnetodielectric, silicon dioxide (SiO_2).**

I. Introduction

SILICON dioxide SiO_2 is a promising material that has been widely studied for applications in microelectronic and optoelectronic devices. The surface coating of silicon dioxide that grows on silicon substrate is hugely beneficial in microelectronics. Because it can protect the silicon, store charge, and block current in electrical applications and even act as a controlled pathway to allow small currents to flow through a device. Another attraction of SiO_2 is its applications on microelectronics, because of its high chemical stability and dielectric properties. It had been reported that its dielectric permittivities of AlN/NiFe/AlN and Ta_2O_5/Co/Ta_2O_5 thin films can be enhanced by increasing the thickness of magnetic layer [1], [2]. The dielectric properties in a trilayer system under different magnetic fields are investigated in the framework of Ginzburg-Landau-Devonshire theory [3]. The properties of Si/SiO_2 and Co/SiO_2 that include the dielectric permittivity and magnetic properties have been widely studied last decades [4]–[7]. The dielectric constant of SiO_2 is around 4.5. The possible reactions between Co and SiO_2 have been studied and formation of ferromagnetic-insulating interface greatly influences an exchange coupling [8]–[14]. In this investigation, we report an up to roughly 55 of dielectric constant of SiO_2 by inserting an extra cobalt thin film and its dielectric variation can be modulated by the inserted Co layer. Significant dielectric enhancement was found and the Maxwell-Wagner model seems to explain the experimental results in the region of interest [15]. We observe the dielectric spectrum of SiO_2 thin films with a controlled thickness of the Co inserted layer under the frequency range from 150 kHz to 30 MHz. The extrinsic properties of magnetodielectric effect [16]–[18] for SiO_2/Co/SiO_2 multilayer films such as dielectric constant and tunability as a function of frequency are measured by varying the thickness of the Co inserted layer and external magnetic fields.

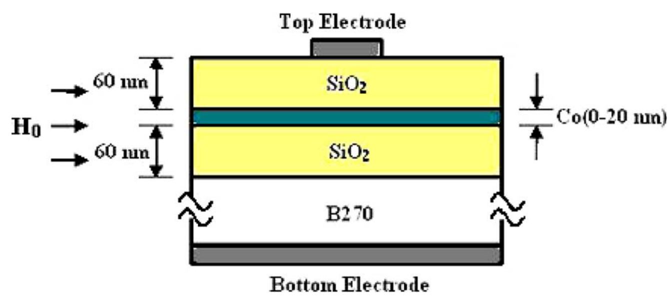

Fig. 1. Schematics for the SiO_2/Co/SiO_2 multilayer films deposited on the B270 glass substrate.

II. Experimental

The multilayered film structures (schematic illustration) in this work were composed of SiO_2(60 nm)/Co (x nm)/SiO_2(60 nm)/B270 glass substrates, as shown in Fig. 1. All films were prepared by ion beam sputtering deposition system at room temperature with a background pressure of 5×10^{-7} Torr. The Si and Co targets were used to fabricate the multilayer films that alternatively deposited the SiO_2 and Co layers on the glass substrates. Before depositing the SiO_2/Co/SiO_2 multilayer films, Si and Co targets were cleaned by ion source for a

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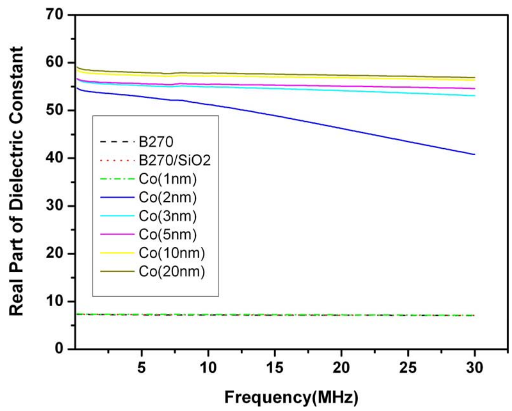

Fig. 2. Permittivity spectrum for the SiO_2/Co/SiO_2 multilayer films without and with different thicknesses of Co inserted layers, respectively.

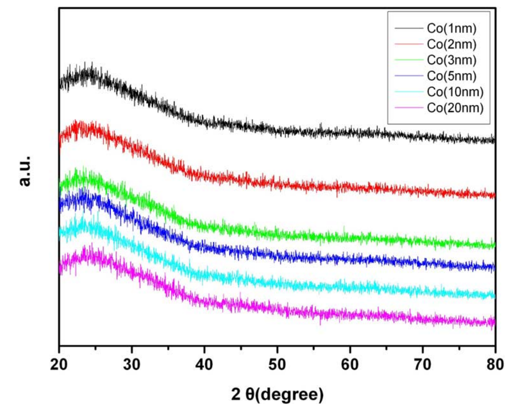

Fig. 3. X-ray diffraction pattern for the SiO_2/Co/SiO_2 multilayer films with 1 nm, 2 nm, 3 nm, 5 nm, 10 nm, and 20 nm thick Co inserted layers, respectively.

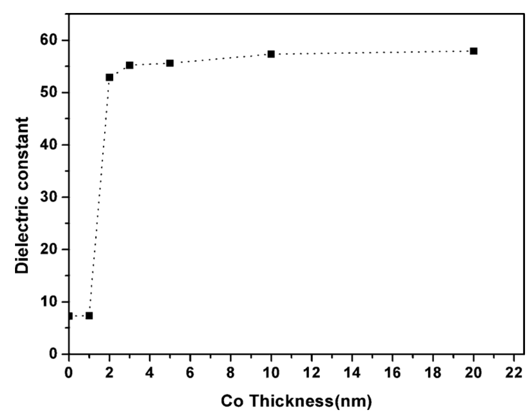

Fig. 4. Dielectric constant variation as a function of the thickness of Co inserted layer for the SiO_2/Co/SiO_2 multilayer films under a frequency of 5 MHz.

few minutes. During the deposition of SiO_2 films, high purity oxygen (O2) 99.999% and argon (Ar) 99.999% were introduced into the growth chamber with the gas composition (1:1) and the pressure was maintained at 10 mTorr to fabricate stoichiometric SiO_2 films. The deposition rates were steadily controlled at around 0.02 and 0.05 nm/s for SiO_2 and Co films, respectively. The dielectric properties for all samples were measured at room temperature with the Agilent 42941B probe and 4294A high precision impedance meters ranged from 150 KHz to 30 MHz with a maximum applied parallel magnetic field of 100 Oe to the film plane. The crystal structure was characterized by ex situ Philips x-ray diffraction (XRD) with Cu Kα radiation. The roughness and cross section thickness of Co/SiO_2/B270 were measured by AFM (DI-3100) and TEM (JEM-2100F). The following analyses will be focused on the variation of dielectric properties for the SiO_2/Co/SiO_2 multilayer thin films with the controlled thickness of Co inserted layer.

III. Results and Discussion

The variation of dielectric constant as a function of frequency range from 150 kHz to 30 MHz for SiO_2/B270 and SiO_2/Co/SiO_2/B270 multilayer thin films with different thicknesses of Co inserted layers is plotted in Fig. 2. It is clearly shown that the dielectric constant is around 7.4 at 150 kHz and slowly decreasing to 7.0 at 30 MHz of the B270 substrate, a single-layer SiO_2 film (120 nm) deposited on the B270 glass substrate (denoted as SiO_2 in whole article), and the SiO_2/Co/SiO_2 thin film with 1nm Co inserted layer. However, the prompt improvement on dielectric permittivity is given by a 2 nm thick Co layer inserted into SiO_2/Co/SiO_2 thin films; i.e., the dielectric constant of the sample with 2 nm Co inserted layer exhibits roughly 55 at 150 kHz and monotonically decreases to roughly 49 at 15 MHz. This takes about 11% of decreasing in dielectric constant. After the thickness of Co inserted layer is thicker than 2 nm, the dielectric constant is roughly between 56.5 and 59 at 150 kHz and monotonically decreases to roughly between 55 and 58 at 15 KHz. Our experimental data indicate that a 2 nm Co inserted layer is the margin of quickly improving the dielectric property of SiO_2 thin films.

The XRD diffraction patterns for the SiO_2/Co/SiO_2 multilayer thin films with the thickness of Co inserted layer varied between 0 and 20 nm are shown in Figs. 3. As it can be seen, no clear diffraction peak is observed. This indicates that the structures for all the SiO_2/Co/SiO_2 samples we fabricated are amorphous structure.

Fig. 4 shows the relationship between dielectric constant and the thicknesses of the inserted Co layer in SiO_2/Co/SiO_2 multilayer thin films under a frequency of 5 MHz at room temperature. The dielectric constant increases with increasing the thickness of the Co inserted layer until it reaches 20 nm. However, it is noted that the dielectric constant of the SiO_2/Co/SiO_2 multilayer thin films increases dramatically roughly at 2 nm, and then only slightly increases when the thickness of Co inserted layer is between 3 and 20 nm under the same frequency. According to the results mentioned above, it indicates that the enhancement of dielectric constant for the SiO_2/Co/SiO_2 multilayer thin films is related to the growth mechanism of the Co inter-layer from island clusters to continuous Co layer between 1 and 2 nm, and this effect is due to the adding of the inserted Co layer in SiO_2/Co/SiO_2 multilayered structures, which redistributes the interface charges between SiO_2 and Co layers. Therefore, the intrinsic polarization is enhanced with increasing its dielectric constant. The enhancement of polarization can be understood by the Maxwell-Wagner polarization, i.e., by inducing mobile

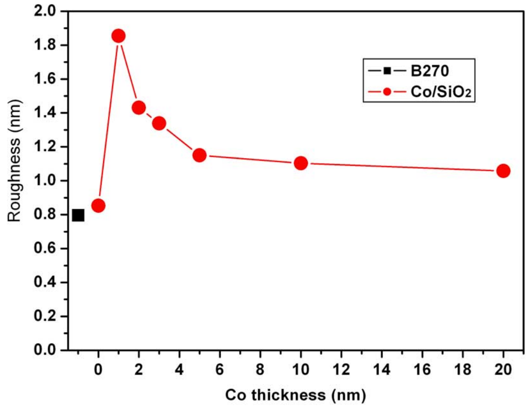

Fig. 5. Root-mean-square (RMS) surface roughness of Co/SiO_2 bilayer films measured by AFM (Taping mode).

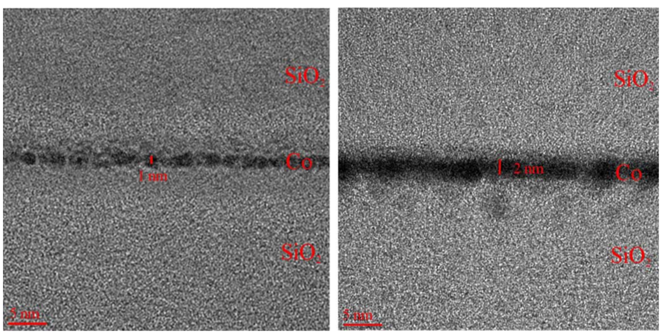

Fig. 6. Cross section images of Co thicknesses(1 nm and 2 nm) in $SiO_2/Co/SiO_2$ trilayer films.

ionic charge in the SiO_2 layers due to the metallic Co inserted layer. In general, interfacial polarization occurs when two adjoining layers differ in conductivity and therefore require different voltage gradients to transconduct a current of constant density. In other words, the Maxwell-Wagner polarization effect is substantial when the relaxation times of the adjacent two materials differ significantly. Therefore, the space charge in the SiO_2 layers can be produced by traveling charge carriers in Co inserted layer. Thus, the piling up of space charge in a volume, or of surface charges at the interface of SiO_2 layer. Due to the Co layer, charge carriers are blocked at inner dielectric boundary layers, leading to a separation of charges which in turn gives rise to an additional contribution to the polarization.

For studying the growth mechanism of the Co inserted layer, we fabricated samples without the top SiO_2 layer; i.e., bilayer samples with Co/SiO_2 structure. Both atomic force (AFM) and transmission electron (TEM) microscopes were used to study the surface morphology roughness (AFM) and cross section images (TEM) of these bilayer and trilayer samples, respectively. Fig. 5 shows the roughness data studied by the AFM. The roughness increases with increasing the thickness of Co layer below 1 nm. After the pick value of roughly 1.85 nm for sample with 1 nm Co, the roughness decreases with increasing the thickness. This is related to the formation of a continuous Co layer on top of the SiO_2 layer after the thickness of Co layer is larger than 2 nm.

The TEM measurement is a good technique for us to study the continuous situation of the Co inserted layer between the SiO_2 layers. As an example, Fig. 6 shows the cross section TEM images of the sample with 1 nm and 2 nm Co inserted layer. The

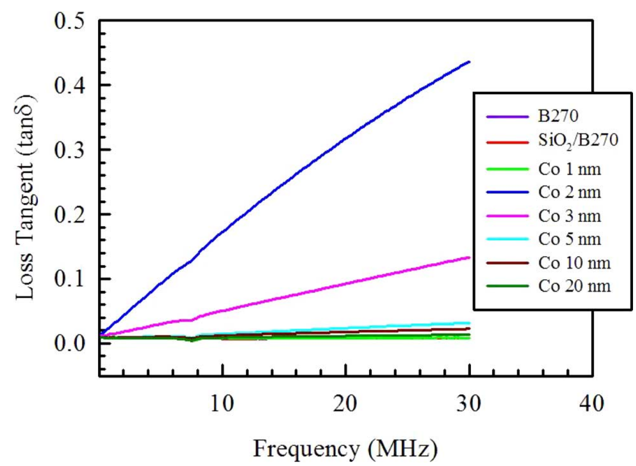

Fig. 7. Loss tangent as a function of frequency of Co inserted layer for $SiO_2/Co/SiO_2$ trilayer films.

dark areas in Fig. 6 are related to the Co element. It is clearly demonstrated that the Co inserted layer with 1 nm is not a continuous layer, i.e., it shows many Co cluster islands between the SiO_2 layers. From both the AFM and TEM studies, we can related the largely increase of the dielectric constant with the formation of a continuous Co inserted layer for samples with the thickness of Co layer larger than 2 nm. The enhancement of dielectric constant in dielectric materials is clearly demonstrated due to the continuous condition of the metallic Co inserted layer between the SiO_2 layers.

The dielectric loss is the power lost in a microwave due to heating as a wave passes through a specific material. The dielectric loss is often expressed as dielectric loss tangent, and low loss makes for a good dielectric while high loss is an absorber or poor dielectric. The loss tangent as a function of frequency of Co inserted layer of all the $SiO_2/Co/SiO_2$ multilayer thin films is depicted in Fig. 8. It is clearly shown that the value of the loss tangent for samples with 2 and 3 nm Co inserted layer is very large, and it is almost more than one order of magnitude larger than that of the other samples. This can be understood due to the leakage current for samples with 2 and 3 nm Co inserted layer is much larger than that of the other samples. For samples with Co thickness larger than 3 nm, the SiO_2 film is cut by the Co inserted layer into two completely separated dielectric layers of SiO_2, therefore, the leakage current of the dielectric layers is reduced again.

For comparing the variation of loss tangent for high frequency application, Fig. 8 plots the loss tangent as a function of thickness of Co inserted layer for the $SiO_2/Co/SiO_2$ multilayer films under a frequency of 1, 5, 10, 20, and 30 MHz. It is clear that for high frequency application, the thickness of Co inserted layer should be larger than 5 nm.

The cobalt is a typical ferromagnetic material and possesses a soft ferromagnetic property. Using the impedance probe and an external magnetic field and the setup as shown in Fig. 1, the dielectric spectrum relative to an external magnetic bias is picked up by the impedance meter. Before the experiment, the $SiO_2/Co(1\ nm)/SiO_2$ thin films is preliminarily measured by the impedance meter sweeping the external magnetic field from 0 to

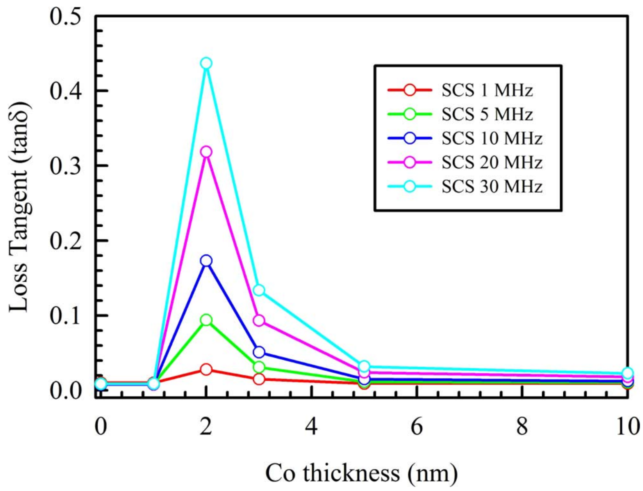

Fig. 8. Loss tangent variation as a function of the thickness of Co inserted layer for SiO_2/Co/SiO_2 trilayer films.

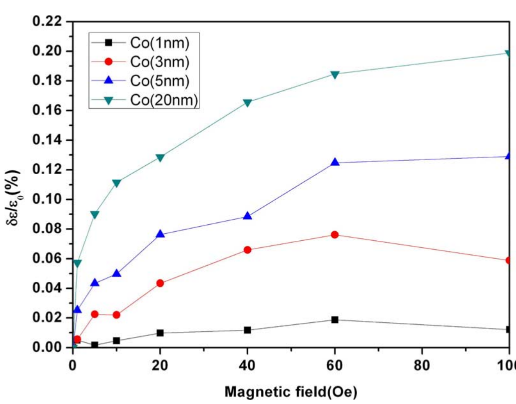

Fig. 9. Relationships of the external magnetic induced dielectric permittivity in SiO_2/Co/SiO_2 thin films with different thicknesses of Co inserted layers.

100 Oe under a frequency of 15 MHz. As can be seen in Fig. 9, the SiO_2/Co (1 nm)/SiO_2 thin film layer demonstrates a nearly constant dielectric property and cannot be changed by the external magnetic bias. The SiO_2/Co/SiO_2 multilayered samples with the thickness of Co inserted layer more than 1 nm are also examined by the same setup and the same process. We have shown experimentally that the SiO_2/Co/SiO_2 films encounter a dielectric variation. The SiO_2 film with Co inserted layer has a 0.04–0.20% dielectric tunability and the variation is saturated roughly above 60 Oe as shown in Fig. 9. This effect can be qualitatively understood by the Ginzburg-Landau-Devonshire theory as discussed in [3]. Here, we have experimentally shown that even with a 3 nm Co inserted layer, the dielectric tunability can be greatly enhanced. Results from the SiO_2/Co/SiO_2 samples with an inserted Co layer of 5 to 20 nm thick conclude that the magnetodielectric phenomena are dominated by the inserted thin soft ferromagnetic Co layer.

IV. Conclusion

In this study, we have experimentally demonstrated that an inserted thin Co layer can enhance the dielectric constant of the SiO_2/Co/SiO_2 multilayered structures. Even with a 2 nm thick Co layer, the dielectric constant can increased roughly from 7.4 to 55. This large enhancement behavior of the dielectric constant could be explained due to the growth mechanism of the Cobalt inserted layer from island clusters to continuous Co layer for samples with x between 1 and 2 nm. For the magnetic induced dielectric variation, the variation of the dielectric constant increased with thickness of Co for samples with x larger than 3 nm. However this increase behavior is roughly saturated for applied magnetic field roughly above 60 Oe. This magnetic tunability of the dielectric constant is clearly attributed to the Co layer in the films. From this study, the magnetodielectric properties are manifested in SiO_2/Co/SiO_2 films and it has the potential for a ferroic sensor application.

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