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Thickness dependence of dielectric properties in sub-nanometric Al₂O₃/ZnO laminates

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In recent years, laminates consisting of alternating layers of two binary oxides with layer thicknesses below 1 nm have attracted attention for their high dielectric constant values, reaching values of about 1000 in the case of Al₂O₃/TiO₂ sub-nanometric laminates. This excellent dielectric performance of the sub-nanometric laminates relies on the Maxwell Wagner (MW) relaxation, exploiting the blocking of the mobile charge carriers of the semiconducting TiO₂ at the interface with Al₂O₃. In this work, we explore the possibilities of enhancing the dielectric constant by MW relaxation in amorphous sub-nanometric laminates of Al₂O₃/ZnO. We demonstrate that the sublayer thickness and the interface of individual layers define the apparent dielectric constant of the laminates. In addition or further understanding, simulations and equivalent circuit analysis of the sub-nanometric laminates were conducted.

Keywords- Maxwell Wagner Effect; laminates; dielectrics; Equivalent circuit

I. INTRODUCTION

The scaling of highly integrated semiconductor devices and problem of quantum mechanical tunneling through thin classical SiO₂ layer triggered the necessity of replacing SiO₂ with a high dielectric constant (k) material. Many binary oxides have been investigated to accomplish the following requirements – lower leakage current, good reliability, chemical stability with the semiconductor [1]. As the primitive requirements are a high dielectric constant (k) and a high bandgap (for low leakage current), materials like HfO₂, ZrO₂, La₂O₃, and TiO₂ have been identified and studied extensively. Single layers of these materials show typically a high k, but the major concerns arise because of their unstable interface with the semiconductor [2] and high dielectric losses [3,4].

In recent years, it has been shown that laminates of an oxide with high bandgap and a semiconducting oxide containing charge carriers can introduce Maxwell – Wagner type polarization, if the individual layer thickness of the laminate is below 1 nm [5,6]. The charges present in the semiconductor accumulate at the interface with the insulating oxide and behave like dipoles, travelling from one interface to the other under application of the *ac* electric field. Many studies have shown that sub-nanometric laminates of Al₂O₃/TiO₂ can show very high dielectric constants (~1000) because of the presence of the Maxwell-Wagner (MW) effect at the interface of Al₂O₃ and TiO₂ layers [8,9].

Even more, it has been reported that this MW polarization can be tailored based on the thicknesses of the insulating and semiconducting layers [9]. As the MW polarization is closely related to space charge polarization, changing the thickness of semiconductor layer, i.e. the distance between the semiconductor/insulating interfaces, the relaxation time of the laminate structure can be tuned which determines the cut-off frequency for the device. Similarly, by changing the thickness of the insulating layer, the dielectric losses can be tuned. Furthermore, the low temperature growth and amorphous characteristics of these laminates allow for their easy integration in electronic devices and current technologies. The goal of this study is to present and evaluate different approaches in order to reduce the losses and increase the working frequency in Maxwell-Wagner laminates.

Though the thicknesses of the individual layers play an important role, another influence on the properties of the laminate is related to the band alignment between the constituent materials. This band alignment has to allow for a sufficient potential difference, so that the charges are confined in the semiconducting layer and long-range transport through the laminate is inhibited. However, knowledge on the insulating gap of Al_2O_3 and TiO_2 in the sub-nanometric thickness range is lacking. Therefore, we have decided to use instead of TiO_2 another binary oxide known for the introduction of charges: ZnO. ZnO has a dielectric constant of 8.6 and a band gap of 3.6 eV [3], and the highly tunable charge carrier concentration induced by oxygen vacancies provides an additional aspect of tailoring the electrical properties of the $\text{Al}_2\text{O}_3/\text{ZnO}$ laminates. But even more, the band gap of ZnO increases when its thickness decreases in $\text{Al}_2\text{O}_3/\text{ZnO}$ laminates due to quantum confinement effect [9,10,11], which could reduce the dielectric losses in sub-nanometric laminates of $\text{Al}_2\text{O}_3/\text{ZnO}$.

Therefore, in this letter we report results of dielectric properties of $\text{Al}_2\text{O}_3/\text{ZnO}$ sub-nanometric laminates for different thicknesses of individual layers of Al_2O_3 and ZnO. We are showing the systematic study of the $\text{Al}_2\text{O}_3/\text{ZnO}$ sub-nanometric laminates constituting the individual layer thicknesses from 0.2 nm to 1 nm. Experimental results and simulations of both dielectric constant and dielectric losses are compared. The electrical properties of the interface between the $\text{Al}_2\text{O}_3/\text{ZnO}$ were determined by simulating the equivalent circuit for the sub-nanometric laminate structure using different dielectric models[13][14].

II. Experimental:

In order to well control the properties of the individual layers in the laminates, the deposition of amorphous single layers of Al_2O_3 and ZnO on Si (001) substrates was carried out. The Pulsed Laser Deposition (PLD) technique has been selected because of the possibility of a good stoichiometry and the thickness control. Deposition was carried out

using a KrF excimer laser with a wavelength of 248 nm. The laser is focused on targets of Al_2O_3 and ZnO with an energy density of about 2 J/cm^2 and a repetition rate of 5 Hz. As in PLD, the deposition parameters play a critical role in defining the properties of the films, the film properties were optimized using different growth parameters (temperature, pressure, laser energy, substrate – target distance and film thickness) for Al_2O_3 and ZnO individually. Based on the optimization, the deposition parameters (temperature – 100°C , oxygen pressure – 0.005mbar, laser energy – 200mJ, target-to-substrate distance – 4.5cm) were selected to develop the amorphous $\text{Al}_2\text{O}_3/\text{ZnO}$ sub-nanometric laminates. The amorphous nature of both Al_2O_3 and ZnO layers was confirmed by the absence of any diffraction peak originating from the laminate in X-ray diffraction θ - 2θ scans. The deposition rates were calibrated by a group of three laminates, where different bilayer thickness were obtained by varying the number of pulses on each target in a systematic way (1000/1000, 1000/100 and 100/1000). The total thickness of the calibration laminates is deduced from X-ray reflectivity (XRR) measurements (Bruker D8 with Cu $K\alpha$ radiation of $\lambda = 1.5406 \text{ \AA}$). Results are presented in figure 1(a). The bilayer Bragg peak, visible for each calibration laminate and indicated by an arrow in the figure 1(a), demonstrates the presence of the artificial periodic arrangement of the Al_2O_3 and ZnO layers throughout the laminate. The bilayer peak in $\text{Al}_2\text{O}_3/\text{ZnO}$ (1000/100) (1000 pulses on Al_2O_3 and 100 pulses on ZnO) laminate represents the periodic layers of 2.5 nm thick Al_2O_3 and 0.4 nm thick ZnO. This indicates that even in the sub-nanometric thickness range, the periodicity of individual layers have been maintained. The deposition rate of ZnO and Al_2O_3 from the calibration laminates were found to be 0.003 nm/pulse and 0.002nm/pulse, respectively.

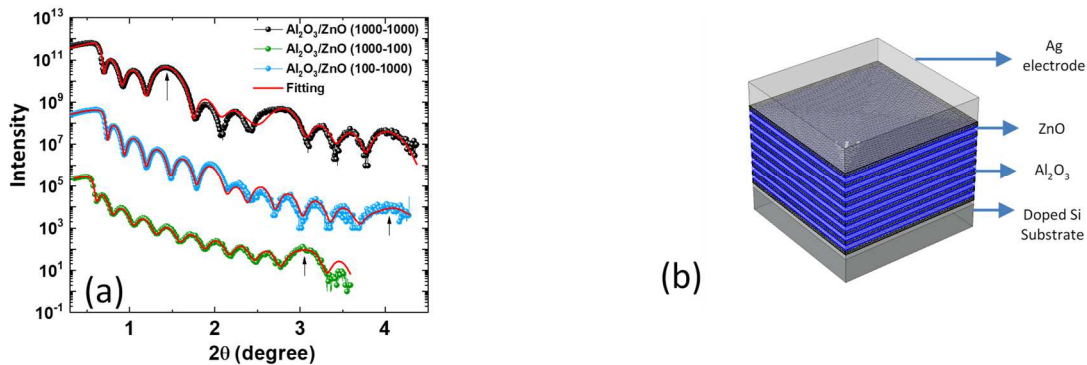


Figure 1 (a) XRR reflectivity curves for calibration samples, black arrow indicate the position of the Bragg peak related to the bilayer periodicity in each sample. (b) Schematics of the laminate structure of Al_2O_3 and ZnO with top electrode (Ag) and the bottom electrode (doped Si).

Based on the XRR curve fitting, a SiO_2 layer ($\sim 1\text{nm}$ thickness) was observed at the substrate/laminate interface. For the growth of oxides on a Si wafer, this SiO_2 layer is virtually unavoidable, and it may have an influence on the overall dielectric properties. A thin SiO_2 layer is also known to reduce the electrically activated defects at the

interface of Si-Al₂O₃, which will further reduce the probability of the charge flow through the interface [15]. Test samples allowing to vary the nature of the interface and the thickness of this oxide layer have shown a negligible influence on the dielectric properties of the laminates, thus the main effect is arising from the internal interfaces of the laminate.

For the measurement of the dielectric properties, capacitor structures with an area of $250 \times 250 \mu\text{m}^2$ were designed and created on the film surface by optical lithography with the lift-off method. The top electrode material is Ag, deposited by thermal evaporation with a thickness of around 100 nm. The Si substrate was used as the bottom electrode. Dielectric properties were measured using an Agilent 4284A precision LCR meter in the frequency domain ($10^2 - 10^5$ Hz).

III. Results and Discussion

For each sample, the apparent dielectric constant and the losses are measured as a function of the frequency (from 10^2 to 10^5 Hz). In Figure 2, the dielectric properties of the Al₂O₃/ZnO laminates for different individual layer thickness configurations (0.2, 0.5 and 1 nm) are drawn. In horizontal direction, the thickness of ZnO layer is increasing while in vertical direction, the thickness of Al₂O₃ layer is increasing. With this series of samples, it is possible to identify the effect on the dielectric properties of the sub-nanometric laminates of (i) the ZnO layer thickness at constant Al₂O₃ thickness (horizontally), (ii) the Al₂O₃ layer thickness at constant ZnO thickness (vertically), and (iii) the total bilayer thickness (diagonal from bottom left to top right). Note that, increasing the total bilayer thickness by keeping the total laminate thickness constant corresponds to a decrease of the number of interfaces in the laminate.

First, we consider the lower Al₂O₃ layer thickness. As we can see, when the thickness of ZnO layer increasing from 0.2 to 1 nm with a constant Al₂O₃ layer thickness of 0.2nm (Fig 2 (7) – (9)), the dielectric constant of the laminate increases and reaches a maximum value of 211 at 200 Hz in the Al₂O₃/ZnO (0.2 nm/1 nm) configuration (Fig 2 (9)). In order to understand the effect involved, the results of these laminates at 200 Hz were compared with the expected dielectric constant of two models: (i) a solid solution of Al₂O₃ and ZnO: In this model, we assumed that, instead of a laminate structure, a solid solution of the constituent materials has been formed and the total dielectric constant of the solid solution was calculated by the volume fraction weighted sum of the single layer dielectric constants. (ii) a series capacitance model: Here, the apparent dielectric constant was modeled by adding the reciprocal capacitance of the different individual layers, but without allowing for a Maxwell-Wagner effect. For the different

models, the dielectric constants of Al_2O_3 of 9.8 and ZnO of 110 were determined from a respectively 40 nm and 25 nm thick single film at 200 Hz deposited under the same growth conditions as the laminates (data not shown here).

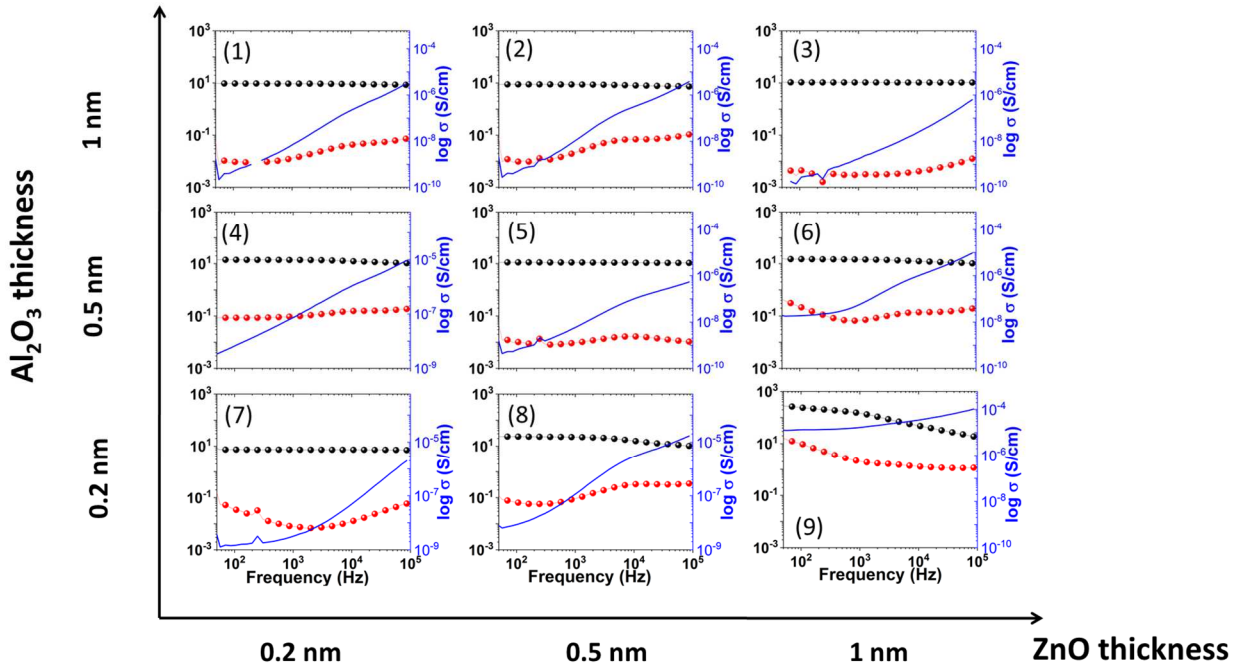


Figure 2 Dielectric constant and loss diagram of laminates with various sublayer thicknesses, maintaining the total thickness of 30 nm. The x-axis is the ZnO individual layer thickness and y-axis the Al_2O_3 individual layer thickness. For example, the bottom left graph represents the results of the $\text{Al}_2\text{O}_3/\text{ZnO}$ (0.2nm/0.2nm) laminate. The black dots represent the dielectric constant and red ones the dielectric losses (left axis). The blue line shows the ac conductivity (right y-axis). The numbers in the diagrams provide the references used in the text and in Figure 3.

Figure 3 shows the comparison of the experimental data of the laminates with the results of these two models, as a function of the ZnO volume fraction, i.e. the ratio of the total thickness of the ZnO individual layers and the total thickness of the laminate. It can be observed that the experimental dielectric constant of the laminates does not follow the solid solution model, confirming the layered structure of all laminates, and especially of the ones with the smallest individual layer thickness, $\text{Al}_2\text{O}_3/\text{ZnO}$ (0.2 nm/0.2 nm). Even for such small values, the layered structure is maintained electrically. For only a small interdiffusion at the interfaces, a dielectric constant corresponding to the solid solution $\text{Al}_2\text{O}_3 - \text{ZnO}$ of 50% would be expected, i.e 59 (Figure 3) due to the high dielectric constant of ZnO. The experimental data follows rather the series capacitance model, except for the $\text{Al}_2\text{O}_3/\text{ZnO}$ (0.2 nm/1 nm). Therefore, the Maxwell-Wagner effect becomes important only for this last laminate, while the other ones behave as expected on the basis of the capacitance of the individual layers. For the $\text{Al}_2\text{O}_3/\text{ZnO}$ (0.2 nm/1 nm) laminate, the two models predict a dielectric constant of 93 and 41, respectively, while the experimentally obtained value is 211. This difference indicates the presence of MW effect enhancing the dielectric constant of the laminate. In fact, the

accumulated charges at the interface of Al_2O_3 and ZnO act as electrical dipoles on application of the alternating electrical field, enhancing the space charge polarization and therefore the apparent dielectric constant of the laminates. At low frequencies, the effect is most prominent, with a relaxation peak in the dielectric losses at moderate frequencies. In the higher frequency range (approximately 10^5 Hz), the dielectric constant decreases to a value corresponding to the solid solution of the laminate composition, as expected in the picture of a charge space polarization.

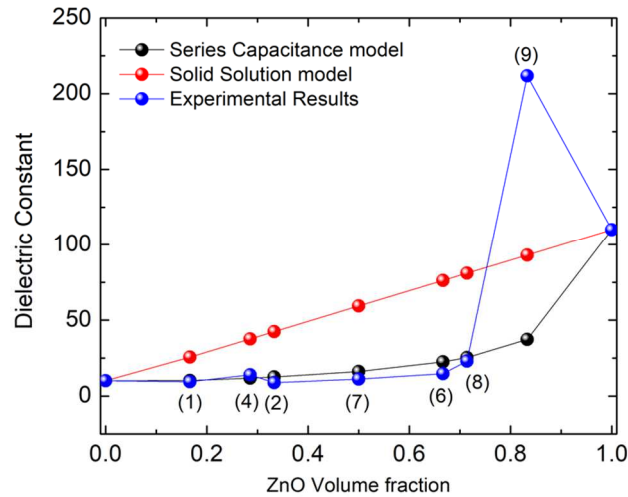


Figure 3 Dielectric Constant of laminates as a function of the ZnO volume fraction, A comparison of experimental data with series capacitance and solid solution model at 200 Hz (the numbers correspond to the different diagrams in Figure 2)

In the range of the ZnO volume fraction between 0.3 and 0.7, the laminates show even a slightly smaller value of the dielectric constant as predicted from the series model, as for example the laminate $\text{Al}_2\text{O}_3/\text{ZnO}$ (0.2nm/0.5nm). Such a decrease of the dielectric constant could be a sign of the introduction of a third element other than the Al_2O_3 and ZnO layer, i.e. the interface becomes electrically active. This would be expected in the case of a MW effect with a low amount of accumulated charges at the interface due to the small ZnO layer thickness. The relaxation frequency of the $\text{Al}_2\text{O}_3/\text{ZnO}$ (0.2nm/0.5nm) sample is in the range of 10^4 Hz, which is higher than the relaxation frequency observed in $\text{Al}_2\text{O}_3/\text{ZnO}$ (0.2 nm/1 nm) in the range of 10^3 Hz. This difference in relaxation frequency could be related to the thickness of the ZnO layer, as the distance between the interfaces decreases (given by the ZnO thickness), increasing the relaxation time and decreasing therefore the relaxation frequency [16].

Furthermore, we can observe that, when the thickness of Al_2O_3 increases from 0.2 to 1 and the ZnO thickness remains constant at 1 nm (Figure 2 (9), (6) and (3)), a gradual decrease in the dielectric losses is observed. This

behavior indicates that a thick layer of Al_2O_3 provides a higher potential barrier to the charge flow from ZnO layer, which is not provided by a thinner Al_2O_3 layer having probably a higher defect density or even pin holes. Laminates with Al_2O_3 thickness of 0.5 and 1 nm (for all measured ZnO thicknesses), show a dielectric constant of about 10, indicating the absence of free charge carriers or an insufficient charge density for MW effect. Although the relative volume fractions of the two constituent materials of the laminates (i.e. the ratio of the total thickness of the layers of each individual material and the total thickness of the laminate) changes, the apparent dielectric constant stays roughly constant. This is to be expected in the framework of the series capacitance constituted by each individual layer of the laminate (see Figure 3). Due to the relatively low dielectric constant of the Al_2O_3 layer in comparison to that of the ZnO layer, the Al_2O_3 layers with constant thickness govern the series capacitance of the laminate, explaining the experimental observation. However, the decrease of the charge density in the ZnO layers, as concluded from the absence of the MW effect, may also indicate chemical effects going on at the interfaces of the laminates, for example an exchange of oxygen between the materials.

Moreover, regarding the number of interfaces in the laminates, i.e. the diagonal of Figure 2 (diagrams (3), (5) and (7)), the dielectric constant stays constant. Thus, we can conclude that MW relaxation is absent in these laminates, in fact the apparent dielectric constant being dominated by the Al_2O_3 layers. The losses decrease for thicker individual layers, which can be explained by the increase of the Al_2O_3 layer thickness.

The ac conductivity of the laminates was calculated from the dielectric constant and dielectric losses [17], shown as the blue line in Figure 2. The presence of a high number of mobile charges in $\text{Al}_2\text{O}_3/\text{ZnO}$ (0.2nm/1nm) can be observed by the presence of a low frequency plateau in conductivity, representing the DC component of the conductivity. From these experimental results, it is clear that for a substantial MW effect in the laminates, a high number of charges in the ZnO layers is needed, although it may enhance the losses.

IV. Simulations and equivalent circuit analysis

For a better understanding of the influence of each individual layer on the macroscopic dielectric properties, the complex impedance of the equivalent circuit composed of three parallel R-C circuits connected in series was simulated. One R-C circuit for each element of the laminate, the Al_2O_3 layer (R_1, C_1), the ZnO layer (R_2, C_2) and the interface (R_i, C_i), was used. The resistance R of each layer is related to the charge dissipation component while the capacitance represents its charge storage component. The total complex impedance (Z^*) for the laminate structure can be expressed as:

$$Z^* = n_b \cdot \left(\frac{R_1}{1 + i\omega R_1 C_1} + \frac{R_2}{1 + i\omega R_2 C_2} + \frac{R_i}{1 + i\omega R_i C_i} \right) + (n_b - 1) \cdot \left(\frac{R_i}{1 + i\omega R_i C_i} \right)$$

Where n_b represents the number of bilayers in the laminate structure. For each bilayer, two interfaces are present, except for the top-most layer. This fact is implemented in the second term. We can deduce the real and imaginary components of the impedance and the electric modulus from the above equations. By fitting the simulation data with the experimentally obtained data, the values of the resistivity and the dielectric constant of each individual layer and the interface are obtained, as well as the thickness of each element (Table 1). The thickness of the individual layers are known from the experimental characterization of the laminates and serve as a quality assessment of the obtained fit results.

Figure 4 shows the comparison of the experimental data and the simulation results of the dielectric constant and dielectric losses for the $\text{Al}_2\text{O}_3/\text{ZnO}$ (0.2/1nm) and $\text{Al}_2\text{O}_3/\text{ZnO}$ (0.2/0.5nm) laminates. The comparison of these two laminates is interesting, as the Maxwell-Wagner effect is more prominent in the first, as compared to the second one. The difference of the simulations compared to the experimental data at low frequency (< 200 Hz) are due to a long-range conduction of charges spilling out from the laminate.

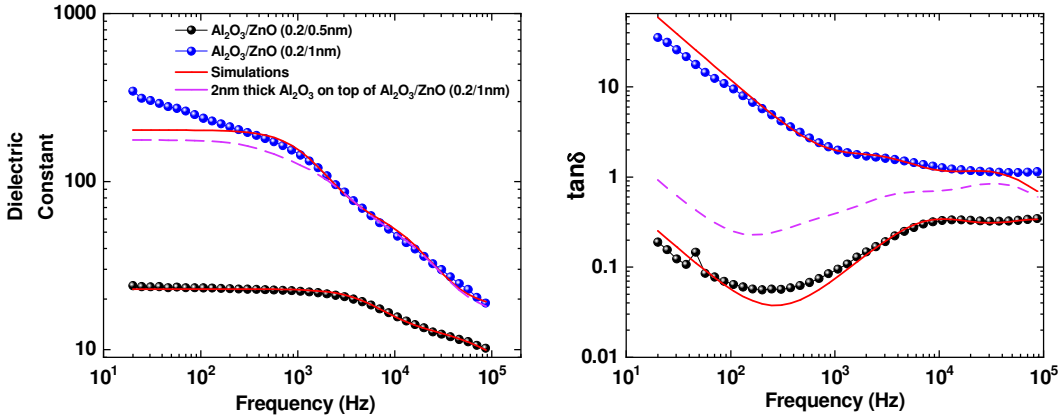


Fig.4 Dielectric constant and dielectric losses of $\text{Al}_2\text{O}_3/\text{ZnO}$ (0.2nm/1nm) and (0.2nm/0.5nm) laminates, experimental and simulation data. Dashed line represent the simulation results for the laminate having an additional 2nm thick Al_2O_3 layer at the electrode/laminate interface

The equivalent circuit simulations of complex samples as the laminates are complicated, and the obtained fit results should be taken with care. However, they allow to deduce some qualitative differences between the two chosen laminates. From the fit results, it is evident that when the thickness of the ZnO layer increases from 0.5 to 1 nm, an abrupt increase in the dielectric constant of the interface is observed, probably due to the accumulation of the charges

at this interface. However, the dielectric constant of the ZnO and Al₂O₃ individual layers is very similar to their bulk values. The resistivity of the interface and the Al₂O₃ layer reduces for a ZnO thickness of 1 nm, indicating that the thinner Al₂O₃ layer is not sufficiently thick to confine the high charge density in 1nm ZnO. Hence, we observe high dielectric losses in the Al₂O₃/ZnO (0.2/1nm) laminate. At the same time, the resistivity of the ZnO layer from 0.5 to 1 nm decreases, indicating a higher charge density and a higher mobility of the charges in a 1nm thick ZnO layer.

The charge density at the interface was calculated by analyzing the capacitance of the interface and the voltage drop at the interface from the equivalent circuit. It was found that the charge density at the interface was 1.49×10^{12} and 1.08×10^{12} cm⁻³ at 200Hz, respectively for the Al₂O₃/ZnO (0.2/1nm) and Al₂O₃/ZnO (0.2/0.5nm) laminate. The higher charge density at the interface of Al₂O₃/ZnO (0.2/1nm) explains the presence of the MW effect and also the high losses through the laminate structure. While in Al₂O₃/ZnO (0.2/0.5nm), a low dielectric constant and a limited MW effect is related to a low charge density at the interface.

In addition, the presented simulations can also be used to calculate the properties of virtual laminates, in order to guide the experimental realizations and optimization of the laminate structure. Again, in the goal to reduce losses, we have carried out the simulation of the properties the Al₂O₃/ZnO (0.2/1nm) laminate adding a 2nm thick Al₂O₃ layer at the laminate/electrode interface (Figure 4). Such an approach was already used experimentally for Al₂O₃/TiO₂ sub-nanometric laminates[18]. The dashed line in Figure 4 shows the simulations which were made on the basis of the values determined by the fitting of the laminate properties and adding one more R-C circuits with parameters corresponding to thick Al₂O₃ (resistivity = 10^8 ohm-m, k = 10). Clearly, the presence of such interface layers could reduce strongly the losses, while limiting the decrease of the apparent dielectric constant.

		Al ₂ O ₃	ZnO	Interface
Al ₂ O ₃ /ZnO (0.2/1 nm)	Resistivity Ω - m	1.9×10^5	1.5×10^4	2.5×10^5
	Dielectric Constant	14	14.81	110
	Thickness (nm)	0.17	0.98	0.15
Al ₂ O ₃ /ZnO (0.2/0.5 nm)	Resistivity Ω - m	1.06×10^9	7.4×10^4	1.6×10^7
	Dielectric Constant	8.05	12.88	19
	Thickness (nm)	0.18	0.45	0.10

Table.1 Fitting results of the Al₂O₃ and ZnO individual layers, and the interface for the simulations shown in Figure 4

(V) Conclusions:

Laminate structures of Al₂O₃/ZnO have been prepared in sub-nanometric range using PLD technique. The XRR measurements were used to calibrate the deposition rates and required laminate configurations were prepared. Impedance spectroscopy was used to determine the dielectric properties of a systematic series of samples with different individual layer thicknesses, allowing to identify the effects of the different layer thicknesses and to compare different models for the interpretation of the results. Due to the presence of MW effect, high dielectric constants of about 200 at 200 Hz were observed in laminates of Al₂O₃/ZnO (0.2/1nm). Moreover, high losses in the films are present due to the high mobile charge density in semiconducting ZnO layer. As the thickness of the ZnO layer decreases in the laminate structure, the charge density at the interface reduces as seen in Al₂O₂/ZnO (0.2/0.5nm).

In addition, simulation studies show that a capping layer of Al₂O₃ at the interface of laminate and electrode can limit the losses through the Al₂O₃/ZnO (0.2/1nm), by applying a layer of Al₂O₃ a reduction in the dielectric losses can be achieved with a limited adverse effect of the high dielectric constant of the film.

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