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Optical and Dielectric Characterization of Atomic Layer Deposited Nb$_2$O$_5$ Thin Films

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The scaling down of future dynamic random access memories (DRAM) requires a dielectric material with high permittivity and low leakage current. Atomic layer deposition (ALD) has been adopted as the deposition technique in microelectronics industry due to its unique self-limited growth mode providing inherent conformality, repeatability and quality of the films. As an alternative to the current ALD techniques, atomic layer deposition of Nb2O5 films has been reported using (tert-butylimido)tris(diethylamido)niobium as the niobium source and ozone as the oxygen source. The effects of deposition and post-deposition annealing conditions, physical thickness as well as the phase composition on the dielectric properties of Nb2O5 thin films have been investigated. In addition, the optical properties of the films have been evaluated. It was found that by tuning the deposition parameters and post-deposition treatments it was possible to obtain high k-values up to 120 with reasonably low leakage current.

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Optical and Dielectric Characterization of Atomic Layer Deposited Nb2O5 Thin Films

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Figure 1 Refractive index at 550 nm for 30 nm thick Nb2O5 films as a function of the deposition temperature.
films and has also been shown to decrease the amount of impurities in the films.\textsuperscript{14}

In order to study the influence of the deposition temperature on the dielectric properties a series of 30 nm thick Nb$_2$O$_5$ thin films was deposited at 250, 275 and 300°C. The corresponding k-values are summarized in Table I. The as-deposited state, the films presented similar k-values (Table I). The effect of the annealing temperature and the resulting phase change from amorphous to orthorhombic Nb$_2$O$_5$ (ICSD card 1840) has also been investigated. The crystallization temperature of Nb$_2$O$_5$ thin films with thicknesses ranging from 15 to 60 nm has been previously determined to be around 550°C.\textsuperscript{10} XRD patterns of 30 nm thick Nb$_2$O$_5$ films deposited at 250, 275 and 300°C and annealed at 550 and 650°C have been recorded (Figure 2a and 2b). When annealed at 550°C, the films deposited at 250°C remained amorphous whereas the films deposited at 275 and 300°C crystallized to the orthorhombic phase. When annealed at 650°C, all the films became crystallized. An increase in the deposition temperature led to a higher peak intensity which indicates higher state of crystallization. A higher deposition temperature favors more complete crystallization and lowers the crystallization temperature. Crystallization upon annealing increased the k-values (Table I).

High k-values ranging from 55 to 105 were measured. The growth of low-permittivity interface layers between Nb$_2$O$_5$ and bottom electrode can lower the permittivity values when using high annealing temperature. The permittivity of the film deposited at 275°C decreased from 105 to 58 when annealed at 550 and 650°C, respectively. Therefore, formation of the orthorhombic phase of Nb$_2$O$_5$ clearly improved the permittivity values of the films but high crystallinity obtained at high annealing temperatures seems to be detrimental to the permittivity value. Therefore deposition temperature of 275°C and annealing temperature of 600°C were chosen for further investigation.

The relation between the thickness and the permittivity of Nb$_2$O$_5$ thin films was investigated. Strong dependence between permittivity and physical thickness (Figure 3) was observed for both as-deposited and annealed films. The drop in the permittivity in thinner films could be explained by a larger effect of the interfaces as compared to the film’s bulk. This behavior exhibited by dielectric thin film, as in the case of STO,\textsuperscript{4,15} has been described as a so called “dead layer” effect.\textsuperscript{16,17} However, this phenomenon is still not fully understood and it is still under investigation whether extrinsic and/or intrinsic effects are provoking it.\textsuperscript{18}

The k-values varied in the range of 18 to 77 and from 25 to 125 for the as-deposited and crystallized films, respectively, when the film thicknesses varied between 13 and 45 nm. We observed that the crystallized films had lower leakage currents than the amorphous films. For example the as-deposited 30 nm thick Nb$_2$O$_5$ film deposited at 300°C had a leakage current density of 10\textsuperscript{-4} A/cm$^2$ at 1 V whereas when annealed the leakage current dropped two orders of magnitude to 10\textsuperscript{-6} A/cm$^2$ at 1 V. It has usually been reported that crystallized films present higher leakage due to current propagation along the grain boundaries.\textsuperscript{19,20} The optical band gaps extracted from ellipsometry measurements present an increase of 0.3 eV for the crystallized Nb$_2$O$_5$ compared to the as-deposited films. This suggests a higher band offset between the high-k Nb$_2$O$_5$ layer and the electrodes, resulting in lower leakage currents for crystallized films.

It should be noted that with lower Nb$_2$O$_5$ film thickness, the leakage current density increases. For example, Nb$_2$O$_5$ films with thicknesses below 25 nm exhibited higher leakage, generally in the order of 10\textsuperscript{-5} A/cm$^2$ at 1 V, even after crystallization. This has been observed in many cases with various high-k oxides.\textsuperscript{21} Possibly, a doping with Al$_2$O$_3$, as in the case of TiO$_2$,\textsuperscript{22} could decrease the leakage current and this work is ongoing.

In summary, this work presents interesting dielectric properties of Nb$_2$O$_5$ films for memory applications. The dielectric properties are sensitive to deposition and post-deposition treatments, and a careful tuning of those for targeted performances is required. Furthermore, the understanding on the influence of the deposition and annealing temperatures on the formation of the orthorhombic phase and degree of crystallization, and their possible influences on the formation of a low permittivity layer, is certainly a key issue for optimization of the dielectric properties of atomic layer deposited Nb$_2$O$_5$ thin films.

Table I. K-values of 30 nm thick Nb$_2$O$_5$ films deposited and annealed at various temperatures.

<table>
<thead>
<tr>
<th>Annealing temperature (°C)</th>
<th>Deposited at 250°C</th>
<th>Deposited at 275°C</th>
<th>Deposited at 300°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>k</td>
<td>As-dep. 550 650</td>
<td>As-dep. 550 650</td>
<td>As-dep. 550 650</td>
</tr>
<tr>
<td>55</td>
<td>42</td>
<td>21</td>
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<tr>
<td>47</td>
<td>105</td>
<td>58</td>
<td></td>
</tr>
<tr>
<td>55</td>
<td>71</td>
<td>67</td>
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</tbody>
</table>

Figure 2 XRD patterns of 30 nm thick films with different deposition/annealing temperatures. Miller indexes are listed for reflections attributable to the orthorhombic phase of Nb$_2$O$_5$ (ICSD card 1840).

Figure 3 Physical thickness effects on the k-values of Nb$_2$O$_5$ thin films as-deposited and annealed at 600°C.
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References