Available online at www.sciencedirect.com
SCIENCE DIRECT®
Microelectronic Engineering 80 (2005) 150–153
MICROELECTRONIC ENGINEERING
www.elsevier.com/locate/mee

Rare-earth scandate single- and multi-layer thin films as alternative gate oxides for microelectronic applications

T. Heeg[1], M. Wagner[1], J. Schubert[1], Ch. Buchal[1], M. Boese[2], M. Luysberg[2], E. Cicerrella[3], J.L. Freeouf[3]

[1] *Institut für Schichten und Grenzflächen ISG1-IT and CNI, Forschungszentrum Jülich*
[2] *Ernst Ruska-Centrum für Mikroskopie und Spektroskopie mit Elektronen / IFF, Forschungszentrum Jülich, D-52425 Jülich, Germany*
Tel: +49 2461 61-5640 email: t.heeg@fz-juelich.de

[3] *Department of Electrical and Computer Engineering, Oregon Health & Sciences University, Beaverton, Oregon 97006, USA*

Abstract

Thin films of rare-earth scandates ($RE$$ScO_3$) as well as multi-layers of scandates and titanates have been prepared using pulsed laser deposition. Epitaxial films were grown on $SrRuO_3$/$SrTiO_3$(100) as well as amorphous films on silicon substrates. The epitaxial films are investigated to measure the physical properties of the crystalline material. Electrical measurements (CV, leakage current) show for example high $\varepsilon_r > 20$ for the scandates and $\varepsilon_r > 35$ for the epitaxial and amorphous multi-layer films. A diffusion of the new materials into silicon is not observed.

Keywords: scandates, titanates, $GdScO_3$, $LaScO_3$, $DyScO_3$, $BaTiO_3$, PLD, multi-layers, laminates

1. Introduction

The continuous structure size reduction in semiconductor technology is leading to a considerable attention for advanced high-κ dielectrics [1]. The rare-earth scandates ($RE$$ScO_3$, where RE is a rare earth element) were recently proposed as candidate materials for the replacement of SiO_2 in silicon MOSFETs in either amorphous or epitaxial form [2]. Epitaxial rare earth scandate thin films have been prepared by pulsed laser deposition technique (PLD) on different substrates. Film stoichiometry and quality were investigated by means of Rutherford backscattering spectrometry (RBS), RBS-channelling, transmission electron microscopy (TEM) and x-ray diffraction (XRD). Electrical measurements have shown a high dielectric constant of around 20 and low leakage currents.

To reach an even higher, tunable dielectric constant while maintaining the favourable properties of the rare earth scandates, multi-layer systems consisting of scandate layers and layers of another

0167-9317/$ - see front matter
doi:10.1016/j.mee.2005.04.058

dielectric material were prepared. This could be a way to benefit from the high dielectric constant of materials as $BaTiO_3$ or $SrTiO_3$ while avoiding their drawbacks (e.g. low stability on silicon, low band offset).

2. Sample preparation

Epitaxial and amorphous $LaScO_3$, $GdScO_3$ and $DyScO_3$ films were grown by PLD using a KrF excimer laser (wavelength 248 nm, pulse width 20 ns and fluence of 2.5 J/cm^2) on different substrate materials. Stoichiometric sintered powder cylinders were used as targets [3]. Samples were prepared in two different target-substrate geometries, the on-axis and the off-axis setup [4]. The substrates were heated by a SiC resistive heater, with direct contact in on-axis setup, and radiation heating in off-axis setup. Deposition took place in an oxygen or nitrogen atmosphere with a pressure of $2\cdot10^{-3}$ mbar in on-axis and $1.2\cdot10^{-3}$ mbar in off-axis setup. For the deposition of multi-layered films, the target material was changed during the experiment, so layers of different materials could be deposited in-situ.

For electrical characterization, circular Pt/Au metal top electrodes structured by standard photolithography/lift-off process and also shadow masking with different areas in the range from 0.00625 to 0.8 mm^2 were finally deposited by e-gun evaporation to form capacitors. For electrical characterization of epitaxial films, $SrRuO_3$ bottom electrodes on $SrTiO_3$(100) substrates also deposited with PLD were used.

3. Properties of *RE*ScO_3 thin films

Structural characterization. Fig. 1 shows an RBS spectrum of an epitaxial $LaScO_3$ film deposited in N_2 atmosphere at 1000°C onto a $SrRuO_3$ bottom electrode layer on a $SrTiO_3$(100) substrate. Simulation [5] shows a good stoichiometry of the film, the La:Sc ratio is 1:1.05, and a film thickness of 150 nm. The sample exhibits a low channelling minimum yield of 3.1% indicating a good crystalline perfection of the sample. Similar results were obtained for $DyScO_3$ and $GdScO_3$ films.

Electrical properties. The capacitance of the epitaxial $LaScO_3$ film is shown in fig. 2. Its value of 140 pF leads to a dielectric constant ε_r of 24. There is no significant dependency from frequency between 10 kHz and 1 MHz. Losses (tan δ) are low, 3.5% at

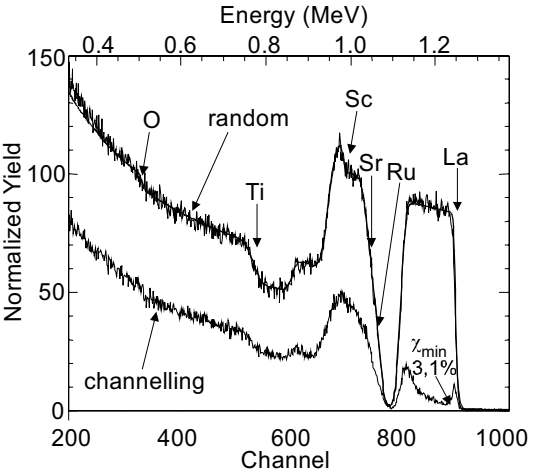

Fig. 1 RBS random spectrum, RBS channelling spectrum and simulation of an epitaxial $LaScO_3$ film on $SrRuO_3/SrTiO_3$.

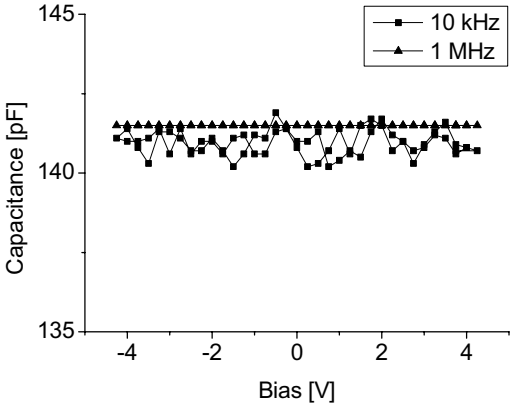

Fig. 2 C(V) of an epitaxial $LaScO_3$ film with Pt/Au top and $SrRuO_3$ bottom electrode on $SrTiO_3$ substrate.

1 MHz and below 1% at 10 kHz. A dielectric breakdown occurs at an electrical field of around 1 MV/cm. $GdScO_3$ and $DyScO_3$ films gave similar results, indicating a slightly smaller ε_r of 20.

The bandgap of the three scandates in amorphous and epitaxial thin films was determined by optical absorption. 1.5 μm films deposited on MgO substrates were used. To enable epitaxial growth of the *RE*ScO_3 on MgO, a thin 10 nm $BaTiO_3$ interlayer was deposited. The results are plotted in fig. 3. The values for the different materials are very similar.

The epitaxial films have an optical bandgap $E_g \approx 5.8$ eV, the value for the amorphous films is lower, $E_g \approx 5.5$ eV. These values are comparable to results obtained by IPE with amorphous films on silicon [6].

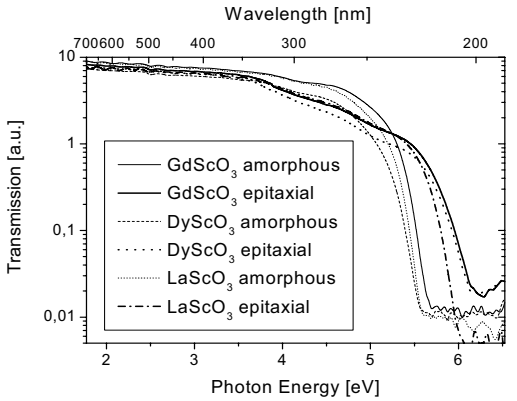

Fig. 3 Optical absorption of amorphous and epitaxial, 1.5 μm thick $GdScO_3$, $DyScO_3$ and $LaScO_3$ films.

4. $BaTiO_3/GdScO_3$ multi-layer stacks

Structural properties. In fig. 4, the RBS spectrum of an amorphous $BaTiO_3/GdScO_3$ multi-layer sample deposited on p-type silicon with IMEC-clean oxide surface is shown. The multi-layer stack consists of 20 double layers of $BaTiO_3$ and $GdScO_3$, the latter deposited first. Simulation indicates a total thickness of 100 nm (5 nm per double layer) and a $BaTiO_3$:$GdScO_3$ ratio of 0.7:0.3. No diffusion into the silicon substrate is visible.

Similar $BaTiO_3/GdScO_3$ stacks were also deposited epitaxially on $SrRuO_3/SrTiO_3$. RBS shows again a 0.7:0.3 ratio of the materials, and a channelling minimum yield of 6%, a low value for such a complex system indicating good epitaxial quality. The TEM image shown in fig. 5 confirms the $BaTiO_3$:$GdScO_3$ ratio measured with RBS. It also underlines the good epitaxial growth of the layers.

Electrical characterization. The epitaxial multi-layer system shows an ε_r of 60 at 1 MHz (film thickness 120 nm, C = 450 pF). It decreases with voltage by 30% at an electrical field of 2.5 MV/cm. No indication of ferroelectricity originating from the $BaTiO_3$ is found. Losses (tan δ) range between 10% and 15%.

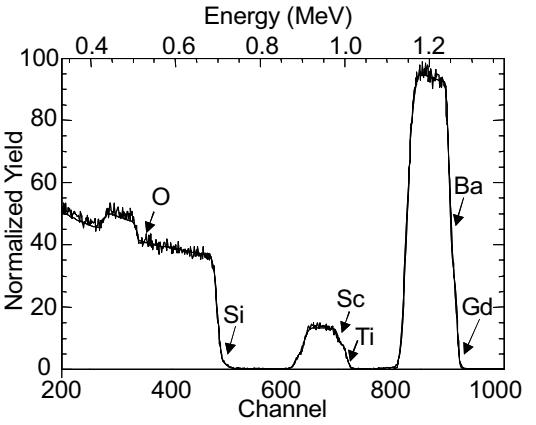

Fig. 4 RBS spectrum and simulation of a 20 double layer stack of $BaTiO_3$ and $GdScO_3$ on silicon substrate.

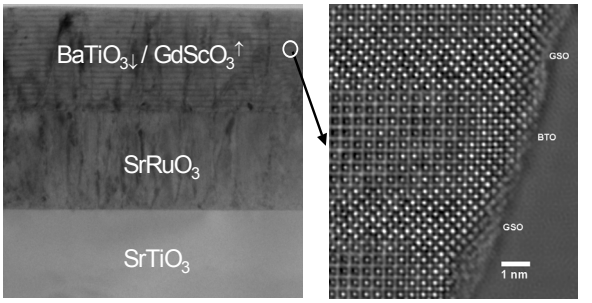

Fig. 5 TEM and high-resolution TEM of an epitaxial 120 nm thick $BaTiO_3/GdScO_3$ stack on $SrRuO_3/SrTiO_3$(100) substrate.

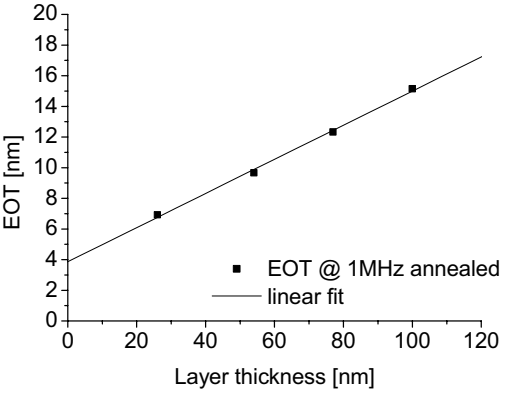

Fig. 6 EOT plot (at 1 MHz) of amorphous $BaTiO_3/GdScO_3$ multi-layer films ranging from 26 to 100 nm.

To determine the electrical properties of the amorphous multi-layer films, the results of several samples with different thickness from 26 nm to 100 nm (5 to 20 double layers) have been evaluated using an EOT plot. The results are shown in fig. 6. The samples were annealed in forming gas at 400°C for 10 min. Au top contacts patterned using a shadow mask were finally deposited. At 1 MHz, the $BaTiO_3/GdScO_3$ system shows an ε_r of 35. The frequency dependency is small, $\varepsilon_r \approx 38$ at 1 kHz. Again, no indication of ferroelectricity is found.

5. Conclusions

Different rare earth scandates were deposited as thin films in epitaxial and amorphous form. The films exhibit promising properties – dielectric constant, band gap, band offset, stability on Si – for the use as alternative gate dielectric. An even higher, tunable dielectric constant can be reached by multi-layered films, in which the scandate fraction maintains the stability on Si and raises the bandgap.

Acknowledgements

This work was supported by the Deutsche Forschungsgemeinschaft (Graduiertenkolleg GRK 549 „Non-centrosymmetric Crystals“).

References

[1] The International Technology Roadmap for Semiconductors, ITRS 2003 Edition, USA (2003) http://public.itrs.net/
[2] D. G. Schlom and Jeffrey H. Haeni, MRS Bulletin **27**, 198 (2002)
[3] Y. Jia, D.G. Schlom, MRI, Penn State University, University Park, 16802 PA, USA
[4] M. Siegert, W. Zander, J. Lisoni, J. Schubert, Ch. Buchal, Appl. Phys. A **69** (Suppl.), S779-S781 (1999)
[5] http://www.genplot.com/RUMP/index.htm
[6] V.V. Afanas'ev, A. Stesmans, C. Zhao, M. Caymax, T. Heeg, J. Schubert, Y. Jia, D.G. Schlom, G. Lucovsky, Appl. Phys. Lett. **85**, 5917 (2004)