# Amorphous High-k LaLuO<sub>3</sub> Dielectric Film for Ge MIS Gate Stack

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### 1. Introduction

A Ge MOSFET with a high-k gate dielectric film is an attractive candidate for high performance devices <sup>[1]</sup>. It has been recently reported that rare earth oxides (RE-oxides) with high permittivity are better for Ge than HfO<sub>2</sub> for Si <sup>[2-4]</sup>. It has been also found that 10at% Ge introduction makes  $Y_2O_3$  amorphous, while it is not the case of HfO<sub>2</sub> <sup>[5]</sup>. Thus, a reasonable gate dielectric film for Ge will be amorphous RE-oxides with high-k. RE-oxides are, however, easily crystallized at relatively low temperatures. In this paper, we propose LaLuO<sub>3</sub> as an amorphous RE-oxide with high-k on Ge, and show its dielectric properties of Ge MIS characteristics.

### 2. Experimental

To raise the crystallization temperature of dielectric films, it is a conventional knowledge that covalent bonding dominated materials such as SiO<sub>2</sub> or Al<sub>2</sub>O<sub>3</sub> are mixed with host materials. There is, however, another way to achieve an amorphous structure. We reported that the crystallization temperature of  $HfO_2$  was increased by mixing  $La_2O_3$  up to 950°C due to a large ionic radius difference between Hf and La <sup>[6]</sup>. This is a guiding principle for making amorphous RE-oxide. In the lanthanoid elements, the ionic radius decreases with atomic number increase due to the lanthanide contraction<sup>[7]</sup>, and La has the largest ionic radius, while Lu has the smallest one. So, the (La2O3+Lu2O3) compound should have a high crystallization temperature than any binary RE-oxides. In fact, LaLuO<sub>3</sub> system has been studied for gate dielectric films on Si [8], but a real advantage of this material on Si has not so far been shown.

We deposited LaLuO<sub>3</sub> by rf co-sputtering of La<sub>2</sub>O<sub>3</sub> and Lu<sub>2</sub>O<sub>3</sub> on HF-last p-Ge(100) wafers, and then annealed (Post deposition Anneal :PDA) in  $N_2$  or  $(N_2+x\%O_2)$ (x :0.001~1) ambient for 30sec from 450 to 600°C. Since the stoichiometry of the deposited film was determined by XPS after the deposition, La(48%)Lu(52%)O<sub>3</sub> and  $La(60\%)Lu(40\%)O_3$  are discussed for  $LaLuO_3$  in this paper. We checked that there was no difference between those films within our experiments. Au and Al were evaporated for gate electrode and the back ohmic contact of MIS capacitors, respectively. The thickness of high-k film was accurately estimated by the combined technique of grazing incident X-ray reflectivity with spectroscopic ellipsometry measurements <sup>[9]</sup>. XRD measurement was used for analyzing the crystal structure. FT-IR and TEM analyses were performed for selected samples. C-V characteristics were measured for MOS samples annealed in various conditions.

# 3. Result and Discussion

The dielectric constant of  $LaLuO_3$  is shown in **Fig. 1** and the k-value is 23 for  $LaLuO_3$ . The experiment was performed on Si with relatively thick  $LaLuO_3$  film to extract the intrinsic dielectric properties of  $LaLuO_3$ . The other

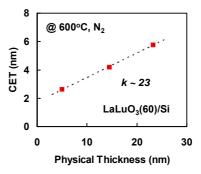


Fig. 1. The relationships between CET and film thickness (determined by GIXR) for 60% La-LaLuO<sub>3</sub>, annealed at 600°C in N<sub>2</sub> ambient. The dielectric constant is estimated from the slope ( $\sim$ 23).

important requirement for the gate dielectric film is the energy band-gap. Fig. 2 shows that  $LaLuO_3$  satisfies the requirements for the energy band-gap of dielectric films (5.5~5.8 eV, measured by the spectroscopic ellipsometry).

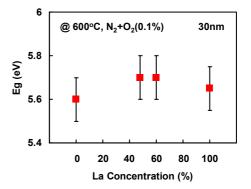
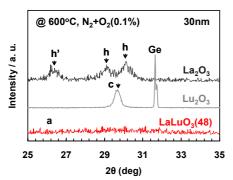


Fig. 2. La concentration dependence of optical band gaps of  $LaLuO_3$  determined by spectroscopic ellipsometry measurements.

Amorphous structure of LaLuO<sub>3</sub> film is really maintained after annealing at 600°C in N<sub>2</sub>+0.1%O<sub>2</sub> ambient as shown in **Fig. 3**, though both La<sub>2</sub>O<sub>3</sub> and Lu<sub>2</sub>O<sub>3</sub> are easily crystallized into hexagonal and cubic phase, respectively. The crystallization temperature was above 850°C. A X-TEM image of **Fig. 4** also shows amorphous LaLuO<sub>3</sub> film throughout the film. Here it should be noted that there is no interface layer between Ge and LaLuO<sub>3</sub> film even after annealing in O<sub>2</sub> ambient. No interface layer such as GeO<sub>x</sub> is also shown in FT-IR measurement, as shown in **Fig. 5**. The interface GeO<sub>x</sub> absorption peak is only observed for the sample annealed in 100%O<sub>2</sub> at 600°C. This will be a significant benefit in terms of the EOT reduction but it gives us a concern about the interface quality.



**Fig. 3.** XRD spectra of 30nm thick 48% La-LaLuO<sub>3</sub>, La<sub>2</sub>O<sub>3</sub>, and Lu<sub>2</sub>O<sub>3</sub> on Ge annealed at 600°C in  $N_2$ +0.1%O<sub>2</sub> ambient (a: amorphous, c: cubic, h: hexagonal, h': hexagonal of La(OH)<sub>3</sub>).

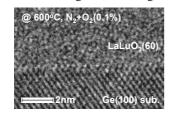
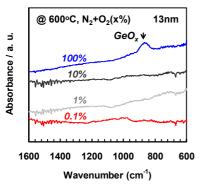
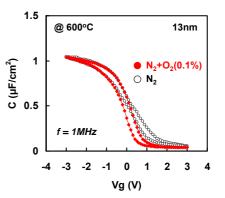


Fig. 4. A X-TEM picture of 60% La-LaLuO\_3/Ge annealed at 600°C in  $N_2{+}0.1\%\,O_2$  ambient.

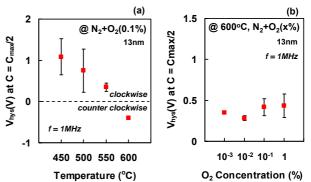


**Fig. 5.** Fourier transform infrared absorption spectra of 48% La-LaLuO<sub>3</sub>/Ge annealed at 600°C in  $N_2+O_2$  ambient with various O<sub>2</sub> concentrations from 0.1% to 100%.

Next, we discuss MIS characteristics of Au/LaLuO<sub>3</sub>/ Ge capacitors. Fig. 6 shows C-V curves at 1MHz of La-LuO<sub>3</sub>/Ge MIS capacitors annealed at 600°C in N<sub>2</sub> and N<sub>2</sub>+0.1%O<sub>2</sub> ambient. The C-V curves are quite good after 600°C annealing and show no difference of saturated capacitance between N<sub>2</sub> and 0.1%O<sub>2</sub> annealed samples thanks to no interface layer Furthermore, a small amount of O<sub>2</sub> significantly improves the interface quality as compared to N<sub>2</sub> annealing. It is really advantageous for the Ge gate stack that the O2-introduced annealing can dramatically decrease the interface states on Ge without any cost of the saturated capacitance (EOT). It is noted that a hysteresis (V<sub>hys</sub>) is not improved in Fig.6 even by O2-introduced annealing. We studied the V<sub>hvs</sub> by changing annealing temperature and oxygen concentration, as shown in Fig. 7. The results clearly indicate that the hysteresis is not originated from oxygen vacancies but from strongly temperature dependent phenomena, and we are optimistic for optimizing the annealing condition from results in Fig. 7.



**Fig. 6.** C-V characteristics of Au/48% La-LaLuO<sub>3</sub>/Ge MIS capacitors annealed at 600°C. The hysteresis is as the counter clockwise, and its window is about 300mV.



**Fig. 7.** (a)  $V_{hys}$  vs. annealing temperature at C=C<sub>acc</sub>/2 of Au/48% La-LaLuO<sub>3</sub>/Ge MIS capacitors annealed at various temperatures in N<sub>2</sub>+0.1%O<sub>2</sub> ambient. (b)  $V_{hys}$  vs. O<sub>2</sub> concentration at C=C<sub>acc</sub>/2 of Au/48% La-LaLuO<sub>3</sub>/Ge MIS capacitors annealed at 600°C with various O<sub>2</sub> concentrations.

#### 4. Conclusions

We have investigated dielectric properties of LaLuO<sub>3</sub> for Ge MIS gate stack. LaLuO<sub>3</sub> has a high dielectric constant (k~23) and amorphous structure after annealed at 600°C. Its energy band-gap is reasonably large (5.5~5.8 eV). The C-V curves annealed in O<sub>2</sub> ambient are quite good and show a high saturated capacitance thanks to no interface layer. It is suggested that the hysteresis in C-V curves can be reduced by optimizing annealing temperature rather than ambient. LaLuO<sub>3</sub> will be a very promising candidate for the high-k dielectric film on Ge.

## Acknowledgements

This work was partly supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology in Japan and partly performed in collaboration with STARC.

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