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Effect of low fluence laser annealing on ultrathin Lu_2O_3 high-*k* dielectric

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The effect of low fluence single pulse laser annealing on a pulsed laser deposited high-*k* dielectric, Lu_2O_3 is reported. With low fluence laser irradiation, high "*k*" of 45 is achieved with an equivalent oxide thickness of 0.39 nm, without taking into account the quantum mechanical tunneling effect. High-resolution transmission electron microscopy micrograph revealed well-ordered epitaxial-like interfacial layer. High-resolution Rutherford backscattering confirmed the presence of Lu-based silicate layer at the interface. It was proposed that the high dielectric constant was caused by the increased ionic polarizability in the film, thereby increasing the ionic contribution of the dielectric constant. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2771065]

The demand for increased performance of Si-based electronic devices to meet the increased complexity in the wireless and information technologies has led to calls for ever decreasing device sizes. According to the semiconductor industry roadmap, the thickness of the silicon dioxide layer will be reduced to 1.2 nm in the year 2013.[1] With the decreased thickness, silicon dioxide is not a suitable material to be used due to the high leakage current, in which the gate current density will exceed the limit set by power dissipation considerations. Rare earth oxides, such as La_2O_3, Yb_2O_3, and Lu_2O_3,[2–5] are promising candidates for the next generation of high-*k* gate insulators which are expected to be thermally stable in contact with Si even at high temperatures. Previously, Lu_2O_3 was reported to have a *k* value of 11–12 (Refs. 3 and 5) with a reported leakage current of 1.2×10^{-3} A/cm^2 (at +1 V) for a 4.5 nm thick film.[3] Lu_2O_3 is expected to have a good conduction band offset with Si and is a suitable high-*k* dielectric candidate because of the large band gap that is expected of Lu_2O_3 due to the completely filled 4*f* shell of Lu and its unique oxidation number (equal to 3), which avoids mixed Lu oxide stoichiometries with different electronic structures.[5] In addition, the 2:3 metal:oxygen stoichiometry ratio promotes a low charge neutrality level and hence a high conduction band offset at the oxide/silicon interface.[5,6]

Laser annealing has been explored for ultrashallow junction formation[7] and recently used in silicide formation for contact metallization in nanoscale devices.[8,9] On the gate stacks, it has been reported that laser annealing caused severe melting.[10] Rapid thermal annealing (RTA) has been used for postdeposition annealing (PDA) in high-*k* materials with the purpose of improving further the deposited film properties in order to achieve better electrical characteristics.[3] The effect of laser annealing on high-*k* dielectric properties is still not known. In this work, we report the effect of low fluence single pulsed laser annealing on a pulse laser deposited high-*k* dielectric, lutetium oxide (Lu_2O_3), with the aim to explore the possibilities of utilizing laser annealing for high-*k* materials, improving the gate dielectric interfacial quality and its microstructure, leading to enhanced electrical behavior.

The Lu_2O_3 ultrathin films were deposited directly on *n*-type (100) Si substrate using the pulsed laser deposition (PLD) technique at a base pressure of 4.5×10^{-7} Torr. A KrF excimer laser with a wavelength of 248 nm and an energy density of 1.5 J/cm^2 was used. The frequency of the laser was set to 5 Hz. The Si substrates were first cleaned using standard cleaning (SC) 1 and SC 2 solutions of Radio Corporation of America, and then dipped in a 1% HF solution to remove the native oxide. The substrates were loaded into the chamber quickly after the last cleaning step to prevent the formation of SiO_2 interfacial layer. Excimer laser irradiation (wavelength of 248 nm and full width at half maximum of 23 ns) was carried out on the as deposited samples using a Lambda Physik laser generator under continuous purified N_2 purging. Laser fluences of 0.2 J/cm^2 were used to study the effect of laser fluence on the electrical characteristics of the dielectric film. Top electrodes of Au with a diameter of 0.3 nm were sputtered using a physical mask. The bottom of the Si substrates was first cleaned using 1% HF to remove native oxide before sputtering it with gold to make the back contact. The film thickness and interfacial property were examined using high-resolution transmission electron microscope (HRTEM) with JEOL 2010 microscope. The electrical characteristics of the fabricated metal-insulator-semiconductor devices were measured using a precision *LCR* meter (HP4284A) at a high frequency of 100 kHz for capacitance-voltage measurements (*C-V*) and semiconductor parameter analyzer (Keithley 4200) for the current density-voltage (*J-V*) measurements. For the high-resolution Rutherford backscattering spectroscopy (HRBS), a beam of 500 keV He^+ is used. The beam was incident on the laser annealed Lu_2O_3/Si sample which was mounted on a high-precision five-axis goniometer installed in a UHV chamber. The incident angle of the beam is 59.5°. The HRBS spectrum is observed at a scattering angle of 65°. According to the simulation of laser interaction with materials,[11] a 0.2 J/cm^2 laser fluence is approximated to generate a surface temperature close to 500 °C in the Si substrate. The characteristics of the Lu_2O_3 film will be discussed and compared to the

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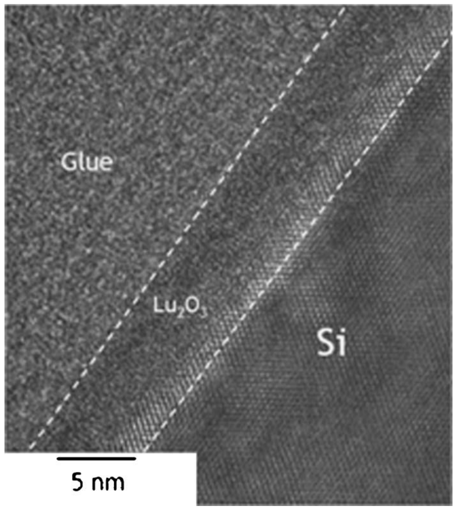

FIG. 1. Cross sectional HRTEM image of Lu_2O_3 thin film after laser thermal irradiation showing an epitaxial behavior at the interface.

samples which were subjected to RTA at 600 °C for 60 s in oxygen ambient, which has shown good characteristics in previous studies reported.[3,4]

Figure 1 shows a cross sectional HRTEM image of the Lu_2O_3 thin film on the Si substrate after laser thermal irradiation process at 0.2 J/cm^2. As observed from the HRTEM image, the thickness of the Lu_2O_3 film is about 4.5 nm. In addition, a crystalline interfacial layer with some degree of epitaxy was revealed in the TEM image. Apart from the interfacial layer, the bulk film appears to be amorphous. The formation of the Lu_2O_3 silicate layer is confirmed by HRBS spectrum shown in Fig. 2. The experimental data was fitted using HRBS simulation software SIMNRA, where the calculated spectrum is shown by the blue line. The HRBS spectrum reveals a two layer stack present in the film, from which a Lu-based silicate layer (~1.0 nm thick) is detected at the interface. This result is close to the representation shown by the HRTEM micrograph in Fig. 1. Previous study on optical properties of Lu_2O_3 revealed that Lu_2O_3 film exhibit weak absorption for wavelength below 400 nm, in which the extinction coefficient is approximated to be around 0.005.[12] The low extinction coefficient translates to an ab-

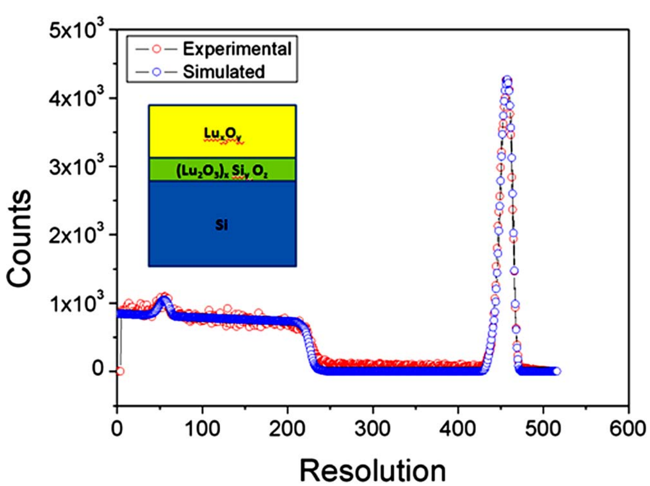

FIG. 2. (Color online) High-resolution RBS spectrum of Lu_2O_3/Si observed at scattering angle of 65°. The inset shows the simulated three-layer stack which corresponds closely to the observed HRTEM micrograph.

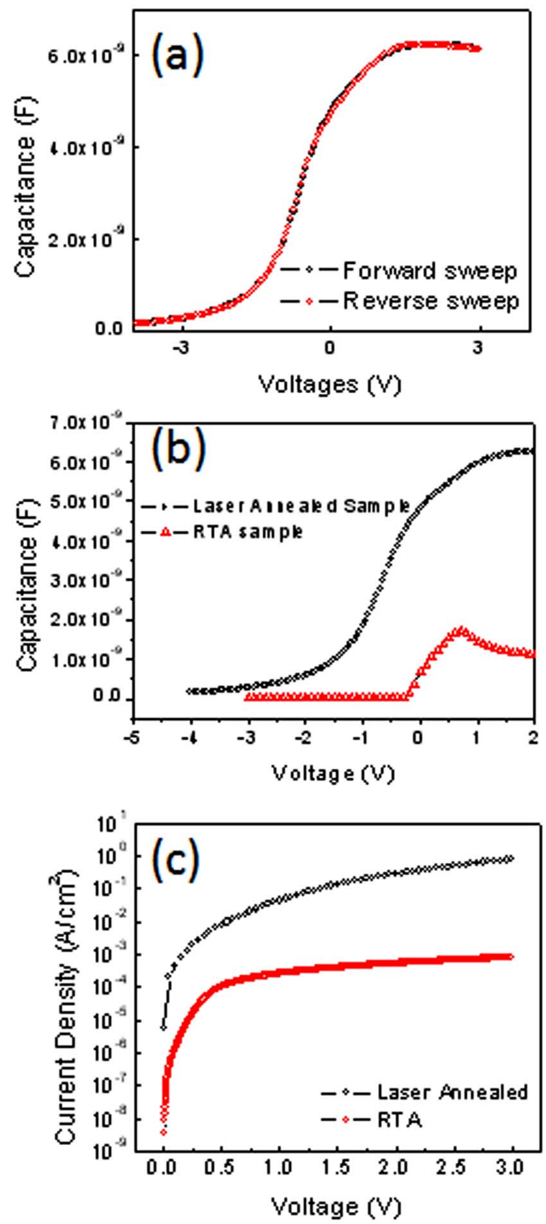

FIG. 3. (Color online) (a) High frequency (100 kHz) capacitance-voltage curve of Au/Lu_2O_3/*n*-type Si capacitors. (b) *C-V* characteristic comparison between laser annealed and rapid thermal annealed samples. (c) Current density-voltage curve of Au/Lu_2O_3/*n*-type Si capacitors of laser annealed and rapid thermal annealed samples.

sorption depth of ~3.94 μm, which is much thicker than our thin film (~5 nm). The thick absorption depth suggests that the heat generated during the laser annealing process is likely to be generated on the Si substrate, rather than in the Lu_2O_3 film itself. The crystalline-epitaxial layer found on the interface is possibly formed via the silicate layer crystallization through local rearrangement of atoms at the interface.

The typical capacitance-voltage measurement at 100 kHz was shown in Fig. 3(a). The curves obtained were well shaped with negligible hysteresis, which suggests minimal interface trap charges in the film. A comparison of the *C-V* characteristic obtained for the sample subjected to rapid thermal annealing at 600 °C in oxygen ambient is shown in the inset of Fig. 3(b). One obvious difference is the accumulation value, with the laser thermal irradiated sample showing about four times higher than the rapid thermal annealed sample. The relative dielectric constant of the laser irradiated and rapid thermal annealed sample is 45.0 and 12.2, respectively. This works out to an equivalent oxide thickness (EOT) of 0.39 and 1.44 nm for the laser annealed and rapid thermal annealed sample, respectively, for the ~5 nm thick samples. The significant improvement in the dielectric constant may be attributed to the epitaxial interfacial layer that is formed after the laser annealing step. One possible contribution of the observed high-*k* value is through the epitaxial crystalline interfacial layer, which was not observed in a typical sample subjected to RTA.[4] For insulating materials, the two major contributions to the static dielectric function are the ionic and electronic contributions.[13] The electronic contribution is usually less than 16. As such, the high dielectric constant should be generated from the ionic contribution. One of the factors affecting the ionic contribution is the ionic polarizability. The larger ionic polarizability relates to a higher ionic contribution, which in turn leads to a higher dielectric constant.[13] It is plausible that the laser thermal irradiation has caused some degree of atomic rearrangement in the film. This rearrangement has led to an epitaxial-like interfacial layer with a highly regular crystallinity and is believed to have caused a higher degree of ionic polarizability which translates to the observed high dielectric constant value. Ionic polarization is induced by the displacement of positive and negative ions with respect to each other in the presence of electric field and is closely dependent on the film density. We found by using x-ray reflectivity (XRR) (not shown) that the density of the laser annealed sample is higher than that of the RTA sample; this suggests that the changes in the film properties have affected the dielectric constant. In addition, XRR data revealed the presence of interdiffused Lu-based silicate layer which corresponded well with the findings using RBS and HRTEM. Improvement of electrical property of ceramic film through laser treatment has been reported elsewhere,[14] where an improved dielectric constant is obtainable after laser treatment. In addition, the observation of the improvement of dielectric property as a result of crystalline epitaxial structure has been observed for Pr_2O_3 in which an effective dielectric constant of 30 is obtainable for Pr_2O_3 grown by molecular beam epitaxy.[15] This is an improvement by a factor of 4, as compared to the amorphous Pr_2O_3 film by metal-organic chemical vapor deposition.[16]

The current density-voltage characteristic (J-V) of the laser thermal irradiated and rapid thermal annealed 4.5 nm thick film is shown in Fig. 3(c). The leakage currents at +1 V bias for the laser thermal irradiated and rapid thermal annealed samples are 4.34×10^{-2} and 2.60×10^{-4} A/cm^{-4}, respectively. One possibility for the increased leakage currents is that the regular arrangement of the interfacial layer is not epitaxial throughout the laser annealed film. It is possible to have regions with different crystal orientations, which may increase the gate leakage current through the grain boundaries and the oxygen vacancy related sites. These grain boundaries act as easy conduction pathways which may become the dominating factors, giving an observed increment of the leakage current density as compared to that of an amorphous nature of the film/interfacial layer. A similar behavior was also observed in the work of Kwo *et al.* where the leakage current characteristic of 4.5 nm thick Gd_2O_3 film with a single domain crystalline has a higher leakage current of about two orders of magnitude as compared to amorphous Gd_2O_3.[17] Despite the higher leakage current, the leakage current density for the laser annealed sample meets the maximum acceptable leakage current listed in the 2006 International Technology Roadmap for Semiconductor for low-standby power devices with similar projected EOT.

In summary, the Lu_2O_3 film subjected to laser thermal irradiation shows good electrical properties in terms of a low EOT of 0.39 nm and a high dielectric constant of 45.0. The leakage current density obtained for the laser irradiated sample is higher than that for the rapid thermal annealed sample; but it is still acceptable for a device with such low EOT. From the optical properties of Lu_2O_3, the heat generation through laser annealing is likely to occur on the Si substrate and therefore the crystalline-epitaxial layer is possibly formed through silicate layer crystallization of the atoms at the interface. It was proposed that the high dielectric constant was caused by the increased ionic polarizability in the film, thereby increasing the ionic contribution of the dielectric constant. Laser thermal annealing provides useful alternative postdeposition treatment in rare-earth dielectric films which may bring about further improvements in gate dielectric applications.

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