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Extended Abstract : Interfacial Layer Formation Mechanisms of $PbTiO_3$ Thin Films Grown on p-Si Substrates by Metalorganic Chemical Vapor Deposition

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Rapid advancements in films growth technology have made possible the fabrication of metal-insulator-semiconductor (MIS) structures with SiO_2, Si_3N_4, Al_2O_3, and $BaTiO_3$ insulator gates. In recent years, the growth of epitaxial insulator films on Si is particularly attractive due to many promising applications such as Si on insulator structures and three-dimensional circuits, and is also important because of their use as a buffer layer prior to the growth of YBa_2Cu_{37-x}. More recently, ferroelectric thin films have attracted attention for nonvolative memories and electro-optic devices. Among many ferroelectric materials, lead titanate ($PbTiO_3$) has been particularly interesting for a wide variety of applications such as pyroelectric infrared sensors. The growth of the $PbTiO_3$ by metalorganic chemical vapor deposition (MOCVD) has been reported in recent years; however, the interfacial layer properties between the $PbTiO_3$ insulator gate and the Si substrate has not been investigated yet.

This presentation shows data for $PbTiO_3$ thin films which were grown on p-type (100) Si substrates in the temperature range between 300 and 800 by low-pressure MOCVD. Even though our main purpose was the growth of $PbTiO_3$ epitaxial films on Si substrates, the delicate problems of epitaxial growth are presented in this letter. Auger electron spectroscopy (AES) was performed to characterize the stoichiometry of the grown films, and transmission electron microscopy (TEM) was carried out in order to investigate the atomic structure of the $PbTiO_3$/Si interface. Furthermore, a possible interfacial layer formation for the initial stage of the $PbTiO_3$ films growth on Si substrates is presented on the basis of the results of the AES and TEM measurements.

The β-diketonate complex of $Pb(tmhd)_2$ (tmhd = 2, 2, 6, 6-tetramethyl-3, 5-heptanedionate), $Ti(OC_3H_7)_4$, N_2O, and argon as the carrier gas were used. The temperatures of the oil baths for $Pb(tmhd)_2$ and $Ti(OC_3H_7)_4$ were maintained at 125℃ and 15℃, respectively. The flow rates of the argon carrier gas for $Pb(tmhd)_2$ and $Ti(OC_3H_7)_4$ were 15 sccm and 10 sccm, respectively. Heating tape was wrapped around the metalorganic-source vapor-transport line, which was at 250℃, to prevent condensation from the source bath to the growth chamber. N_2O gas was supplied through a separate line into a resistively, heated, vertical, cold-wall reaction chamber at a system pressure of 1 Torr.

The composition of the $PbTiO_3$ thin layer and the $PbTiO_3/Si$ interface was investigated by AES. The results showed that the film consisted of lead, titanium, and oxygen at a 50Å depth of the $PbTiO_3$ layer. The ratio of the peak-to-peak intensities among the lead, titanium, and oxygen peaks of the films was similar to that of $PbTiO_3$ grown by chemical vapor deposition.

In addition to AES measurements, TEM measurements were performed to demonstrate the formation of an interfacial layer in the $PbTiO_3/Si$ structures. These measurements were performed on adjacent pieces of the same samples which were used for the AES measurements. A bright-field TEM image was clearly observed that there was an interfacial layer between the top $PbTiO_3$ layer and the bottom Si substrate. Therefore, the formation of the interfacial layer was confirmed again. The thickness of $PbTiO_3$ was the same as that from the AES measurements. The thickness of the interfacial layer was approximately 100Å which was in reasonable agreement with that obtained from AES measurements. The spots and the ring in the patterns indicated the Si substrate and the $PbTiO_3$ polycrystalline layer, respectively. A high-resolution TEM image of the $PbTiO_3/Si$ structure indicate that an interfacial layer is formed in the $PbTiO_3/Si$ interface and the $PbTiO_3$ thin film is polycrystalline. Even though our main purpose was tha growth of $PbTiO_3$ epitaxial layers on Si substrates, the formation of the polycrystalline $PbTiO_3$ might be considered to be due to the existence of the interfacial layer prior to the growth of the $PbTiO_3$ layer. It is impossible to explain unambiguously from the measurements with certainty why this interfacial layer is form. Even though all metalorganic sources contribute to a complicated growth process, and probably quite complicated, a qualitative idea of the growth mechanism can be suggested.

In summary, AES measurements showed that a $PbTiO_3$ thin film on the Si substrate was grown by low-pressure MOCVD at a relatively low temperature. TEM measurements showed that an interfacial layer was grown by MOCVD and that an interfacial layer was formed between the $PbTiO_3$ and the Si. The interfacial layer formation mechanism between the $PbTiO_3$ and the Si was introduced.