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Initial reactions of Al_2O_3 films on Si(001) grown by atomic layer deposition: in-situ synchrotron radiation photoemission spectroscopy

김석환[1,2], 박호영[1,2], 백재윤[2], 김민국[2], 전철호[2], 안종렬[2], 황찬국[3], 황한나[3], 박종윤[2], 안기석[1]

[1]한국화학연구원, [2]성균관대학교 물리학과, [3]포항가속기연구소

Recently, a scaling-down tendency for electronic devices bring about changes in both deposition technique and material of ultrathin layers. First of all, Atomic layer deposition (ALD), most promising method, has provided good solution for various ultrathin layers with certain advantages such as layer by layer control of the film thickness, uniform roughness, and appropriate chemical stoichiometry. As one of high dielectric materials, Al_2O_3 has attracted interest due to the thermal stability in contact with Si, a large band gap (9 eV), and a high dielectric constant (k=9.8). However, the initial reaction to form a Al_2O_3 layer on Si substrate has not been elucidated in spite of an importance of the interface status in ultrathin film. Therefore, the interface formation should be investigated using ALD method which providing highly uniform films with thickness control to the atomic layer level.

In this study, initial reactions and interfacial characteristics of ultra-thin Al_2O_3 films were investigated with in-situ layer-by-layer deposition and analysis using ALD method and synchrotron radiation photoemission spectroscopy, respectively. At the first, H_2O/Si(001), terminated by -OH and -H, was used as a starting substrate. The chemical compositions and electronic structure at each cycle of ALD processes were analyzed by measuring core-level spectra of Al 2p, O 1s, and Si 2p and valence band spectra, respectively. As a result, core-level spectra reveal that the initial ALD reaction produce a ultrathin interface layer composed of Al_2O_3, Si oxide, and Al-silicate. Al_2O_3 layer increased upon repeating ALD cycle, but small amount of Si oxide and Al-silicate still remain in the interface. On the other hand, valence band offsets between Al_2O_3 layer and Si are saturated to the similar value of Al_2O_3 film (3.72eV) after 3rd cycle. This suggest that ultathin Al_2O_3 insulating layer is formed after 3 cycle of ALD process notwithstanding the formation of Si oxide and Al-silicate.