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論 文 要 旨

THESIS SUMMARY

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要旨（英文 800 語程度）
Thesis Summary (approx.800 English Words)

The increasing computational demands of artificial intelligence (AI) technology are driving advancements in semiconductor research, focusing on developing higher computational efficiency through innovative architectures, novel device operation mechanisms, and new materials exploration. Advanced memory technology with in-memory computing (IMC) unlocks extraneous potential for computational capability and energy efficiency. The integration of non-volatile memory (NVM) technology demonstrates significant promise for realizing IMC and storage class memory (SCM). Within the development of NVM technology, ferroelectric materials have been drawing much attention thanks to their unique charge storage nature. The newly discovered wurtzite III-N ferroelectrics exhibit fascinating properties such as extraordinarily high remanent polarization (P_r) of over 100 $\mu C/cm^2$, low dielectric constant (ε) of 11-20, and good compatibility to CMOS technology. The ferroelectricity of binary III-N compounds is enabled by foreign dopants such as Sc atom, leading to the formation of aluminum scandium nitride (AlScN). The polarization switching mechanism of AlScN is facilitated by Sc content and governed by the translation of N atoms in its wurtzite crystal structure. Despite AlScN thin films feature tunable ferroelectricity by varying Sc doping concentration, less-than-ideal endurance performance (~10^5 cycles) and large leakage are urgent issues that should be dealt with. In this research, fabrication, characterization, and analysis are implemented for AlScN films to further improve the endurance performance by two orders and achieve below 5 V operation.

For capacitor fabrication, this research focuses on *in-situ* reactive sputtering of TiN/AlScN/TiN metal-ferroelectric-metal (MFM) capacitors and dedicates to exploring the deposition conditions for better film quality. Herein, the dependence of sputtering conditions such as Sc content of the AlSc alloy target, process pressure, and target-substrate (T/S) distance are examined. It is found that low Sc content film is more favorable over high Sc film due to wider bandgap (E_g), suppressed leakage, lower ε, higher P_r, and enhanced breakdown field (E_{BD}). Plus, with the assistance of lower process pressure, the sputtered films demonstrate lower leakage and boosted E_{BD}, along with reduced ε_i and increased P_r. Additionally, a suitable T/S distance could also enhance the overall ferroelectricity and E_{BD}. However, a trade-off relationship is found between the coercive field (E_c) and E_{BD}. These results not only paved the way for improved endurance performance but also created the opportunity for thickness downscaling.

In order to tackle the high E_c characteristics of AlScN, defect engineering via oxygen incorporation is applied, and the role of oxygen-related complex defects is analyzed and understood. Intentionally incorporating O_2 gas flux into the Ar/N_2 sputtering ambient results in AlScON films with complex defects that allow *c*-axis shrinkage. Accordingly, E_c decreases along with higher ε. Moreover, this technique breaks the aforementioned trade-off as E_{BD} is improved in AlScON films. Note that the incorporated O atoms distributed nonuniformly inside the film, which allows for partial polarization at low bias, resulting in a comparatively linear P_r gain over a wider range of E. Such performance is feasible and can be harnessed as multi-level operation in analog circuits for AI applications. Besides, with defect engineering and TiN serving as electrodes, this research highlights the demonstration of aggressive thickness scaling down to 9 nm of AlScN with under 5 V operation. Nevertheless, severe fatigue effect in endurance is found due to trapping/de-trapping incidents at Sc-O bonds. Decreasing the Sc content in the film could be helpful to mitigate the fatigue effect. Furthermore, oxygen profiling of multi-layer AlSc(O)N ferroelectrics shows that multi-interfaces in the ferroelectric stack could also help inhibit the leakage path and increase the breakdown strength.

In this research, it is identified that considerably high oxygen concentration exists at the ferroelectric/bottom electrode interface, which could greatly affect the endurance performance with non-negligible fatigue effect. Hence, oxygen depletion at the interface is conducted through sputter deposition in reducing ambient, with H_2 gas flux blending into the Ar/N_2 sputtering ambient to lower the partial pressure of residual oxygen. Though incorporating H_2 gas flow could deteriorate the *c*-axis crystallinity, AlScN films exhibit excellent robustness against reducing ambient as the ferroelectricity sustained up to over 10% of H_2 flux. The decreasing ε serves as a corroborating indicator of the lower oxygen content in the deposited films. It is worth mentioning that with oxygen depletion the leakage is suppressed along with enhanced E_{BD}. This implies a different mechanism that can be explained as E_g broadening. Most importantly, the wake-up and fatigue effects are mitigated by oxygen depletion, showing the effectiveness of oxygen level lowering at the interface. A record high endurance cycle of 2×10^7 switching cycles is achieved, pushing the endurance limits of AlScN thin films.

The originality of this research is to improve the reliability and scalability features of AlScN films through several consecutive steps: (1) sputtering condition exploration, (2) defect engineering via oxygen incorporation, and (3) interface oxygen depletion by reducing ambient deposition. Based on the results, guidelines which describe exquisite combinations of Sc and O content profiling to multi-layered ferroelectric stack are proposed for future high endurance and low-voltage operation. In summation, this research provides insight into enhancing the reliability and scalability of AlScN thin films and hopefully contributes to the development of ferroelectric AlScN for future advanced NVM technology.