9th Asia-Pacific Conference on Plasma Physics, 21-26 Sep, 2025 at Fukuoka

Nucleation-Controlled Sputtering Growth of Epitaxial and Non-Epitaxial Oxide Semiconducting Thin Films

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The initial stage of crystalline film growth plays a crucial role in achieving high-quality crystalline films. In particular, during sputtering deposition, control over nucleation at this stage is critical, as the competition between kinetics and thermodynamics governs the growth process and ultimately influences the structural and functional properties. Here, we demonstrate epitaxial growth of oxide semiconducting films through nucleation control during the early growth stage, focusing on $Zn_{1-x}Mg_xO$ and $(ZnO)_x(InN)_{1-x}$ (ZION) films on sapphire substrates.

$Zn_{1-x}Mg_xO$ is a wide-band-gap semiconductor with high exciton binding energy, strong tolerance to radiation, and the composition based on Earth-abundant elements, etc. Although research into $Zn_{1-x}Mg_xO$ for electrical and optoelectronic devices dates back several decades, recent technological developments have reignited substantial interest in this material. $Zn_{1-x}Mg_xO$ are now being explored as promising platforms not only for optoelectronic devices but also for next-generation nonvolatile memories, such as resistive switching and ferroelectric memories, which are essential for applications from conventional storage to neuromorphic and multilevel computing. However, its integration is hindered by lattice mismatch with common substrate, sapphire (18%). To address this, we developed an "inverted" Stranski-Krastanov (SK) growth method [1,2]. This approach involves the formation of strain-relaxed nanosized 3D islands during the buffer layer growth, which then coalesce into flat 2D layers. Despite the difficulties in forming such nanosized islands due to the surface energy cost associated with their large surface-to-volume ratios, we achieved this through two mechanisms: thermodynamically, by promoting the adsorption of insoluble impurities that lower the surface energy, and kinetically, by increasing the ratio of the deposition rate (DR_{buffer}) to the adatom surface diffusion coefficient, thereby increasing the nucleation density. These islands effectively relaxed the lattice mismatch strain, yielding atomically flat $Zn_{1-x}Mg_xO$ films with a high out-of-plane alignment, as evidenced by the full width at half-maximum (FWHM) of 0002 x-ray rocking curve (XRC) of 72 arcsec (Fig. 1).

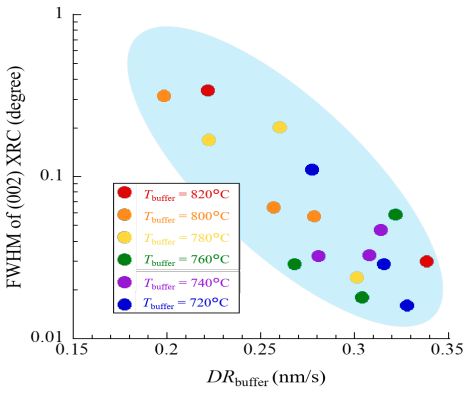

Fig. 1. FWHM of 0002 XRC of inverted SK grown $Zn_{1-x}Mg_xO$ films on sapphire as a function of deposition rate of buffer layer.

We then extended this approach to ZION, a pseudo-binary alloy of ZnO and InN. ZION offers tunable band gaps across the entire visible spectrum, high optical absorption coefficients, and large exciton binding energy [3], making it a promising material for optoelectronic solar energy-harvesting devices. Yet challenges such as low miscibility of ZnO and InN, stoichiometry control of such multinary compounds, and 19% lattice mismatch with sapphire have limited its development. We addressed the first two challenges employing a non-equilibrium magnetron sputtering technique, combined with atomic flux control of O and N via vacuum ultraviolet absorption spectroscopy (VUVAS) [4]. To fully unlock the potential of ZION, we now focus on overcoming the remaining challenge of achieving epitaxial growth on conventional substrates using the 3D ZnO island buffer layers. This enabled the epitaxial growth of high-quality ZION films with a remarkably narrow FWHM of 144 arcsec for 0002 XRC, surpassing those grown on lattice-matched, yet costly, bulk ZnO substrates. Interestingly, we found that superior crystal quality is achieved not only with a higher DR_{buffer} but also with a smaller substrate off-cut angle (θ_{off}). A small θ_{off} ($\leq 0.4°$) promotes the formation of nanoscale 3D islands by maintaining crystallographic terrace widths wider than the adatom migration length, thereby suppressing strained step-flow growth and enabling effective strain relaxation at the island surfaces. These results present a complementary perspective to conventional epitaxial growth approaches, which typically favor lower deposition rates and larger θ_{off} to enhance step-flow growth. This 3D-ZnO buffer layer technique offers a kinetically driven pathway for high-quality epitaxy on lattice-mismatched substrates, not only advancing ZnO-based optoelectronic and energy-harvesting applications, but also providing a scalable method for other materials that lack cost-effective lattice-matched substrates.

References

[1] Itagaki et al., Sci. Rep. 10, 4669 (2020).
[2] Nakamura, et al., Cryst. Growth Des. 22, 3770 (2022).
[3] Itagaki, et al, Mater. Res. Express. 1, 036405 (2014).
[4] Matsushima et al., IEEE Trans. Plasma Sci. 45, 323 (2017).