

5th Asia-Pacific Conference on Plasma Physics, 26 Sept-1Oct, 2021, Remote e-conference Sputter epitaxy of ZnO based oxide/oxynitride semiconductors

for excitonic transistors

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Regarding crystalline films growth on large lattice mismatched substrates, there are two primary modes by which thin films grow on a crystal surface or interface. They are Volmer-Weber (VW: island formation) mode and Stranski-Krastanov (SK: layer-plus-island) mode. Since both growth modes end up in formation of three dimensional (3D) islands, fabrication of single crystalline films on lattice mismatched substrates has been challenging. Here, we add another growth mode, inverted SK mode (island-plus-layer growth mode) [1]. In this mode, relaxed 3D islands initially grow with the help of impurities that reduce the surface energy, and the islands rapidly coalesce to form 2D layer after desorption of impurities. Eventually, films grow in 2D mode and form single crystals. This crystal growth via 3D-2D mode transition, opposite to 2D-3D transition in the well-known SK growth, is what we call inverted SK mode. This new mode enables us to grow single crystalline films even on large lattice mismatched substrates. Here, we demonstrate growth of single crystalline ZnO films on 18%-lattice mismatched sapphire substrates via the inverted SK mode, where nitrogen atoms are employed as impurities. Furthermore, we report a new class of ZnO based materials, $(ZnO)_x(InN)_{1-x}$ (hereafter ZION) [2, 3], which are synthesized on the single-crystalline ZnO films. ZION have tunable band gaps across the entire visible spectrum, high exciton binding energy (30-60 meV), and high optical absorption coefficients (10⁵ cm⁻¹), all of which make ZION promising materials for excitonic devices such as exciton transistors.

All films were fabricated by radio-frequency magnetron sputtering. First, we fabricated ZnO films via inverted SK growth mode. 10-nm-thick buffer layers consisting of 3D islands were initially deposited on c-plane sapphire substrates at 735°C in Ar/N_2 atmosphere. Then, 1000-nm-thick ZnO films were deposited on the buffer layers at 700°C in Ar/O_2 atmosphere. Finally, 100-nm-thick ZION films were fabricated on the single crystalline ZnO films. The ZION films were deposited at room temperature in $Ar/N_2/O_2$ atmosphere using ZnO and In targets.

We have succeeded in growth of single crystalline ZnO films on 18%-lattice-mismatched sapphire substrates via inverted SK mode. Here, buffer layers consisting of nano-sized 3D islands initially grow with the help of adsorbed N atoms on the film surface. After cessation of N-atom supply, the islands rapidly coalesce to form 2D layers where an increase in the surface energy provides a driving force for coalescence. Finally, the films grow in 2D mode, forming atomically flat surface with steps of 0.26 nm in height. We found that there are two important parameters for the growth of high-quality films through the inverted SK mode: 3D island diameter and the surface roughness of the buffer layer. In a small island (<10 nm in diameter) with a large surface-to-volume ratio, the strain relaxation mainly occurs not inside but at the surface of the island, resulting in a low dislocation density and thus a low mosaicity. Buffer layers consisting of such islands promote grain coalescence during the subsequent growth if the surface is smooth enough for adatom migration. Furthermore, we have succeeded in fabrication of single crystalline ZION films on the ZnO films. Figure 1 (a) shows the atomic force microscopy (AFM) image, indicating that the ZION film has atomically flat surface and a step-terrace structure. From x-ray reciprocal space mapping around (105) plane (Fig. 1(b)), we found that the a-axis lattice parameter of the film is same as that of ZnO, that is, the film grows coherently on the ZnO films.

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References

- [1] N. Itagaki, et al., Sci. Rep., 10, 46691 (2020).
- [2] N. Itagaki, et al., Mater. Res. Express. 1, 36405 (2014).
- [3] N. Itagaki, et al., U.S. Patent, No.8274078 (2012).

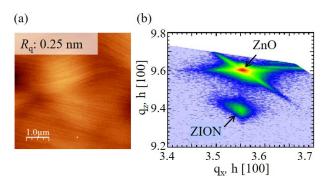


Figure 1. AFM image (a) and reciprocal space map around (105) plane (b) of a 100-nm-thick ZION film fabricated on a ZnO film grown via inverted SK mode. Here R_q denotes root mean square roughness of the surface.