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Sputter epitaxy through “inverse” SK mode for a new class of excitonic devices

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Regarding crystalline film 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 (Fig. 1a). Since both growth modes end up in formation of threedimensional (3D) islands, fabrication of single crystalline films on lattice mismatched substrates has been challenging. Recently, we found another growth mode, “inverse” SK mode (island-plus-layer growth mode) (Fig. 1b) [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 inverse SK mode. This new mode enables us to grow single crystalline films even on large lattice mismatched substrates. Here we report growth of single crystalline ZnO films on 18%-lattice mismatched sapphire substrates through the inverse SK mode, where nitrogen is employed as impurity. Furthermore, we demonstrate epitaxial growth of $(ZnO)_x(InN)_{1-x}$ (hereinafter referred to as ZION) films using the single crystalline ZnO films as templates. ZION films have tunable bandgaps over the entire visible wavelength range, and the large exciton binding energies of 30-60 meV make ZION promising materials for excitonic transistors [2, 3].

Figure 2 shows the evolution of surface morphology upon deposition of 10–1000 nm of ZnO films. Here, we first deposited 10-nm-thick films with N_2 gas, then cease supplying N_2, and finally deposited 1000-nm-thick ZnO films without N_2. For comparison, an AFM image of a 1000-nm-thick ZnO film fabricated without N2 through all growth stages is shown in Fig. 2f. In the case with N_2, nanosized 3D islands initially grow, and they rapidly coalesce to form 2D layer after cessation of N_2 supply. Eventually, single crystalline ZnO film grows, where the film has atomically-flat surface. In the case without N_2, by contrast, ZnO film grows in a 3D mode through all growth stages. The resultant 1000-nm-thick film has 3D columnar structures, and the root-mean-square (RMS) roughness of the surface is large of 30 nm. These results clearly show that impurities play a key role in determining the growth mode of heteroepitaxy for lattice-mismatched systems.

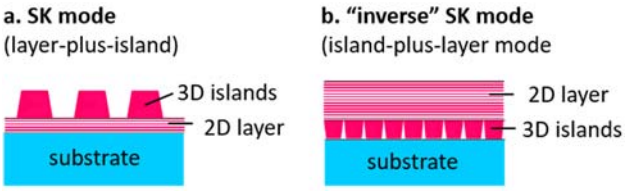

Fig. 1. Schematic diagram of the possible growth modes of heteroepitaxy on large lattice-mismatched substrates: **a** Stranski-Krastanov (SK: layer-plus-island) mode, and **b** island-plus-layer growth mode where a relaxed buffer layer consisting of 3D islands initially forms and a relaxed two-dimensional (2D) layer subsequently grows.

Furthermore, single crystalline ZION films with atomically flat surface have been fabricated using the single crystallin ZnO films as templates. We observed the direct band gaps of ZION, and thus the high absorption coefficient of 10^5 cm^{-1}. Strong blue and green photoluminescence (PL) from ZION films were observed at room temperature, where the films were excited by a cw He-Cd laser. We have also succeeded in the fabrication of ZION/ZnO QWs and optical switching of exciton flux.

We believe that our findings on the sputter epitaxy will open a new pathway for high-quality film growth of wide variety of materials that have no lattice-matched substrates.

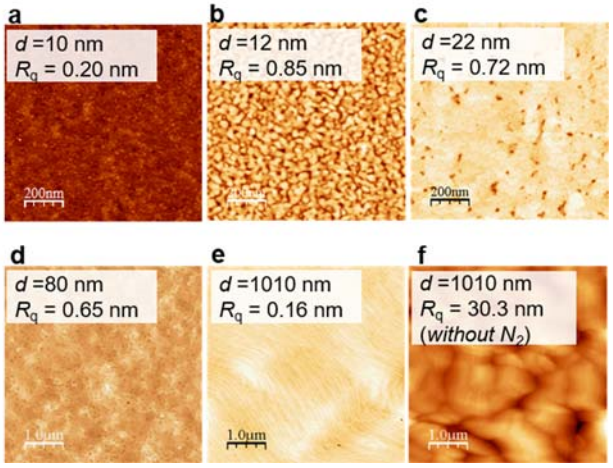

Fig. 2. AFM images showing the evolution of surface morphology upon deposition of 10–1000 nm of ZnO films (**a-e**). N_2 gas was introduced only for the early stage deposition of 10-nm-thick film. An AFM image of ZnO film fabricated without N_2 through all stages is also shown for comparison (**f**).

References

[1] N. Itagaki, et al., *Sci. Rep*. **10**, 4669 (2020).
[2] N. Itagaki, et al., *Mat. Res. Express* **1**, 036405 (2014).
[3] N. Itagaki, et al., *U.S. Patent* No. 827407 (2012).