

2nd Asia-Pacific Conference on Plasma Physics, 12-17,11.2018, Kanazawa, Japan **Sputter epitaxy of high quality (ZnO)**_x(InN)_{1-x}: a new semiconducting material for excitonic devices

Naho Itagki, Nanoka Miyahara, Daisuke Yamashita, Kunihiro Kamataki, Kazunori Koga, and Masaharu Shiratani

Graduate School of Information Science and Electrical Engineering, Kyushu University e-mail : itagaki@ed.kyushu-u.ac.jp

An exciton, which is an electrically neutral quasiparticle, is a bound state of an electron-hole pair attracted by the electrostatic Coulomb force. The most interesting feature of an exciton is that it can be generated by and converted back into a photon within a short time (<nsec). Thus, excitonic devices potentially bring great improvements to the speed of electronic-optical (E/O) conversion along with significant miniaturizations of E/O converters. The major challenges for excitonic devices are finite exciton binding energy (Eex) and finite exciton lifetime. The small exciton binding energy of typical excitonic materials, such as GaAs, limits the device operation temperature below 125K. While, the exciton lifetime in such materials is less than a nanosecond, allowing excitons to travel only a small distance before it recombines. Recently, we have developed an excitonic device with a new semiconducting material, (ZnO)_x(InN)_{1-x} (abbreviated as ZION) [1, 2], synthesized by sputter epitaxy. The large Eex of ZION potentially enables excitonic devices that are operational at room temperature. Another advantage of ZION is the large piezoelectric constant. Since the overlap integral of electron-hole wavefunctions in ZION quantum wells (QWs) can be noticeably small because of the strong piezo-electric field enhancing the spatial separation of electrons and halls [3]. The elongated exciton lifetime should allow excitons in QWs to travel long distance before it recombines.

In this report, we focus on the growth of high-quality ZION films, aiming to reveal the intrinsic nature of ZION as a material for excitonic devices. Fabrication of single crystalline ZION films, however, has been challenging because no bulk crystals of ZION exist, and no lattice matched substrates exist for ZION across almost the entire composition range of $(ZnO)_x(InN)_{1-x}$. In this context, we proposed a new mode of heteroepitaxy on lattice mismatched substrates, "inverse Stranski-Krastanov (SK) mode", in which, high density 3D islands are formed at the early stage of crystal growth [4]. Since the strain induced by the lattice mismatch between the films and substrates is relaxed through the island formation, such 3D islands have low density of misfit dislocation, resulting in good in-plane and out-of-plane alignment of the crystal axis. Then, on 3D islands, crystal grains that grow originating from the 3D islands can be coalesced with other crystal grains, and eventually grow in a layerby-layer fashion (2D growth). Here, we report on the surface morphology, electrical properties, and photoluminescence (PL) of ZION films grown in the inverse SK mode.

ZION films were prepared on 18%-lattice mismatched c-sapphire substrates through 3 steps. First, we prepared ZnO 3D islands of 10 nm by magnetron sputtering. Second, ZnO 2D layers of 1 μ m were prepared on the 3D islands. Finally, ZION films of 30 nm were formed on the ZnO 2D layers. The chemical composition ratio of ZION films was (ZnO)_{0.92}(InN)_{0.08}.

Single crystalline ZION films with atomically flat surface have been successfully prepared via inverse SK mode on 18%-lattice mismatched substrates. Figure 1 shows the atomic force microscopy (AFM) image of ZION films. The film shows a step-terrace structure with the step height of 0.27 nm, which corresponds to the height of a single-atomic-layer step and the half-length of the c-lattice parameter of ZION. We found that there are two key parameters for achieving such inverse SK growth, one is the density of 3D islands, and the other is the surface height distribution of 3D islands. XRD analysis clarified that the lattice distortion drastically decreases with increasing the grain density of 3D layers. This result indicates that dense 3D islands, which have large surfaceto-volume ratios, contribute to efficient release of strain energy at the grain surfaces due to the lattice mismatch between epitaxial films and substrates. Meanwhile, the crystal growth of 2D layers was observed to be very sensitive to the surface height distribution of 3D layers. The smooth surface of 3D layers with homogeneous height distribution enhances the migration of adsorbed atoms, resulting in the subsequent lateral growth of 2D layers. Finally, we observed strong blue PL from the ZION film at room temperature. The inset in Fig. 1 shows the PL image of the ZION film, where a cw He-Cd laser excited the film. Since the no PL emission was observed for polycrystalline ZION films, we considered that grain boundaries provide non-radiative recombination centers. We believe that the high-quality ZION films obtained in this study will open up a new avenue for excitonic devices.



Figure 1. AFM image of a ZION film grown by sputter epitaxy. The inset shows the PL image of the ZION film.

References

- [1] N. Itagaki, et al., Mat. Res. Express 1 (2014) 036405.
- [2] N. Itagaki, et al., U.S. Patent No. 827407 (2012).
- [3] N. Itagaki, et al., Proc. SPIE, 9364 (2015) 93640P.
- [4] N. Itagaki, et al., Opt. Engineering, 53 (2014) 087109.