Temperature-dependent photoluminescence study on aluminum-doped nanocrystalline ZnO thin films by sol-gel dip-coating method

Giwoong Nam[a,*], Hyunggil Park[a], Hyunsik Yoon[a], Soaram Kim[b], Jae-Young Leem[a,b]
[a*]Department of Nano Engineering, Inje University, Gimhae 621-749, Republic of Korea
[b]Department of Nano Systems Engineering, Center for Nano Mab Inje University, Gimhae 621-749, Republic of Korea

초 록: The photoluminescence (PT) properties of Al-doped ZnO thin films grown by the sol-gel dip-coating method have been investigated. At 12 K, nine distinct PL peaks were observed at 2.037, 2.592, 2.832, 3.027, 3.177, 3.216, 3.260, 3.303, and 3.354 eV. The deep-level emissions (2.037, 2.592, 2.832, and 3.027 eV) were attributed to native defects. The near-band-edge (NBE) emission peaks at 3.354, 3.303, 3.260, 3.216, and 3.177 eV were attributed to the emission of the neutral-donor-bound excitons (D0X), two-electron satellite (TES), free-to-neutral-acceptors (e,A0), donor-acceptor pairs (DAP), and second-order longitudinal optical (2LO) phonon replicas of the TES (TES-2LO), respectively. According to Haynes' empirical rule, we calculated the energy of a free exciton (FX) to be 3.374 eV. The thermal activation energy for D0X in the nanocrystalline ZnO thin film was found to be ~25 meV, corresponding to the thermal dissociation energy required for D0X transitions.

1. 서론

In recent decades, investigations for blue and UV photonics have become the focus of interest. The preparation of the first GaN-based light-emitting diode with emission was achieved in 1993. ZnO is a potential competitor of GaN for blue and UV light-emitter and detector application, and also high-power, high-frequency electronic devices. ZnO with a band gap of 3.4 eV has become a natural choice considering its low toxicity and abundance in the earth. ZnO is characterized by a large exciton binding energy (~60 meV), as compared to the 26 meV exciton binding energy in GaN, which allows the stable existence of excitons and deficient excitonic lasing operation at room temperature or even higher. The sol-gel process in combination with the dip-coating process offers the greatest possibility for low-cost small and large-area coating of ZnO thin films for technological applications. Despite extensive research over the past several years, some fundamental properties of the low-temperature photoluminescence (PL) in ZnO by sol-gel dip-coating are still not fully understood.

2. 본론

Figure 1 shows the PL spectrum of the nanocrystalline ZnO thin film at low temperature (12 K) and the draft of the anticipated level of defect in the nanocrystalline ZnO thin film. At 12 K, nine distinct PL peaks were observed at 2.037, 2.592, 2.832, 3.027, 3.177, 3.216, 3.260, 3.303, and 3.354 eV. The deep-level (DL) emissions (2.037, 2.592, 2.832, and 3.027 eV) were attributed to native defects. The peaks at 2.037 and 3.027 eV were attributed to interstitial oxygen (Oi) and a zinc vacancy (VZn), respectively. We supposed that the peak at 2.832 eV was probably due to a high exciton binding energy of 60 meV or interstitial aluminum (Ali). In Fig. 1(b), the peak at 2.592 eV was generated by VZn and above proposal. The near-band-edge (NBE) emission peaks at 3.354, 3.303, 3.260, and 3.216 eV were attributed to the emission of D0X, TES, (e,A0), and DAP, respectively. The other peak at 3.177 eV, which correspond to the TES-2LO are obtained from the fittings.

Figure 2 shows the temperature dependence of the PL spectra for nanocrystalline ZnO thin film between 3.10–3.40 eV within a temperature range of 12–180 K. In general, the D0X emission is dominant at low temperatures. In particular, because of the abundance of D0X in ZnO, the photon emissions FX and D0X are usually mixed. As the temperature increases, the emission of FX becomes dominant, and localized carriers are thermalized to occupy higher energy states, which leads to a blueshift in D0X, TES, (e,A0), and DAP. This blueshift behavior is observed in the temperature range 12 K < T < 50 K. With increasing temperature, the DAP emission energy was redshifted in the temperature range 50 K < T < 180 K.

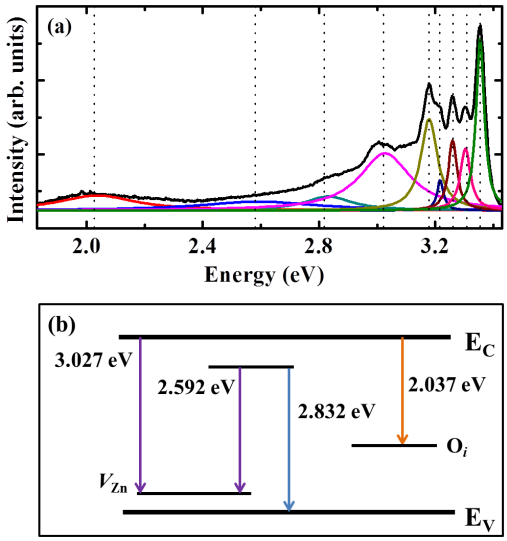

Fig. 1. (a) PL spectrum of the nanocrystalline ZnO thin film at 12 K and (b) the draft of the anticipated level of defect in nanocrystalline ZnO thin film.

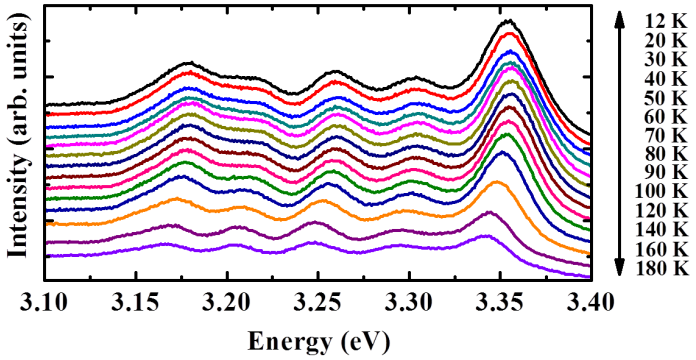

Fig. 2. Temperature-dependent PL spectra of NBE emissions of nanocrystalline ZnO thin film grown on quartz substrate.

3. 결론

We report temperature-dependent PL studies on Al-doped ZnO thin films grown by the sol-gel dip-coating method. At 12 K, nine distinct PL peaks were observed at 2.037, 2.592, 2.832, 3.027, 3.177, 3.216, 3.260, 3.303, and 3.354 eV. According to the peak positions of the TES and D0X, we calculated ED as 68 meV. According to Haynes' empirical rule, we calculated the energy of FX as 3.374 eV. The temperature-dependent D0X transition energy was fitted by Cody's equation. The experimental data were also fitted with Eg(0) = 3.354 eV, k = 0.18 eV, and θ = 500 K. The thermal activation energy for the D0X in the nanocrystalline ZnO thin film was found to be ~25 meV, corresponding to the thermal dissociation energy required for the D0X transitions.

참고문헌

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