Yellow electroluminescence from sputtering synthesized aluminum nitride nanocomposite thin film containing aluminum nanocrystals

M. Yang*, T. P. Chen*, Y. Liu** and J. I. Wong*

* School of Electrical and Electronic Engineering, Nanyang Technological University
Singapore 639798, yangming@pmail.ntu.edu.sg
** State Key Laboratory of Electronic Thin Films and Integrated Devices
University of Electronic Science and Technology of China, Chengdu, Sichuan, P. R. China 610054

ABSTRACT

In this work, visible electroluminescence (EL) from aluminum nitride (AlN) thin film containing aluminum nanocrystals (nc-Al) prepared by a radio frequency sputtering technique is reported. The yellow EL shows a spectrum peaked at 565 nm. A linear relationship between the EL and the current transport in the nc-Al/AlN nanocomposite thin film is observed, and both the current transport and the EL intensity show a power-law dependence on the gate voltage. The current conduction is explained by the carrier conduction in the percolation networks of tunneling paths formed by the nc-Al arrays, and the light emission is attributed to the radiative recombination of the injected electrons and holes via the deep-level defects at the locations of nc-Al along the tunneling paths. The EL intensity is also temperature dependent. The enhancement in EL intensity at the elevated temperatures is observed and associated with the enhanced current conduction. At both room temperature and elevated temperatures, there is no obvious decay in the EL intensity for up to 10^6 on/off cycles, indicating the excellent light emission endurance. The results in this work suggest the potential applications of the nc-AlN/AlN nanocomposite thin film in the low-cost and CMOS-compatible optoelectronic devices.

Keywords: aluminum nitride, aluminum nanocrystal, electroluminescence, light emitting device

1 INTRODUCTION

Aluminum nitride (AlN) exhibits outstanding physical properties, such as wide bandgap, high thermal conductivity, and good match of both the thermal expansion coefficient and the lattice constant to a Si substrate [1]. Recently, the sputtering synthesized AlN nanocomposite thin film containing Al nanocrystals (nc-Al) has been reported, and its non-volatile memory applications and carrier transport behaviors have been demonstrated [2][3]. In this work, we extend the application of such nc-Al/AlN thin film to the optoelectronic applications. We demonstrate that a visible electroluminescence (EL) in yellow color can be obtained at room temperature and elevated temperatures from the light emitting structure based on the nc-Al/AlN thin film. Excellent light emission endurance can also be achieved. Due to the low-cost of synthesis and the compatibility to modern complementary metal-oxide-semiconductor (CMOS) process, the nc-Al/AlN thin film has the potential application in CMOS-compatible optoelectronic devices.

2 EXPERIMENTAL DETAILS

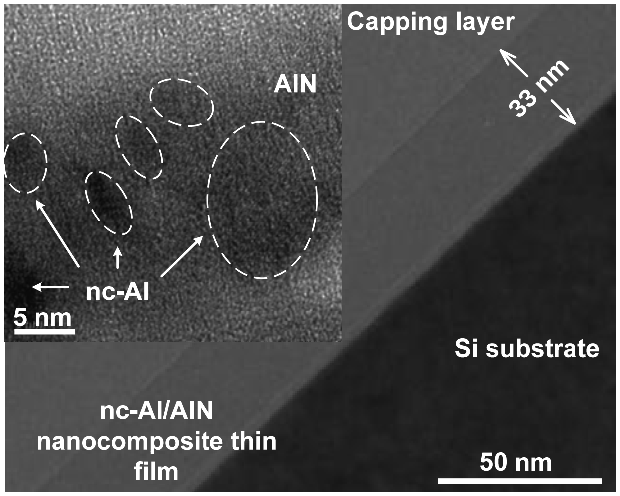

Figure 1. Cross-sectional TEM image showing the nc-Al/AlN nanocomposite film. The inset shows the existence of nc-Al embedded in the AlN matrix.

The nc-Al/AlN nanocomposite thin film was fabricated on <100> *p*-type Si substrate by the radio-frequency (rf) magnetron sputtering of pure Al target (99.999%) in the gaseous mixture of Ar and N_2. Before the deposition, both the Al target and the Si substrate were pre-sputtered to remove the contamination. During the deposition, the flow rate ratio between Ar and N_2 was fixed at 1:3 and the rf power was 500 W. The previous x-ray photoemission spectroscopy (XPS) analysis confirms the as-deposited AlN thin film is Al-rich [2]. The cross-sectional transmission electron microscope (TEM) image of the nc-Al/AlN nanocomposite thin film is shown in Figure 1. The nc-Al embedded in AlN matrix with a size of less than 5 nm can be observed. The light emitting device based on the nc-Al/AlN thin film was formed by the deposition of indium tin oxide (ITO) gate electrodes and the Al backside contact. A Keithley 2400 semiconductor characterization system was used for both the current-voltage and EL measurements. The light emission measurement was carried out with a photomultiplier tube (PMT) detector and a monochromator. The endurance measurement was done by the pulse generator unit of Keithley 4200 semiconductor characterization system. All the measurements were conducted in dark environment.

3 RESULTS AND DISCUSSIONS

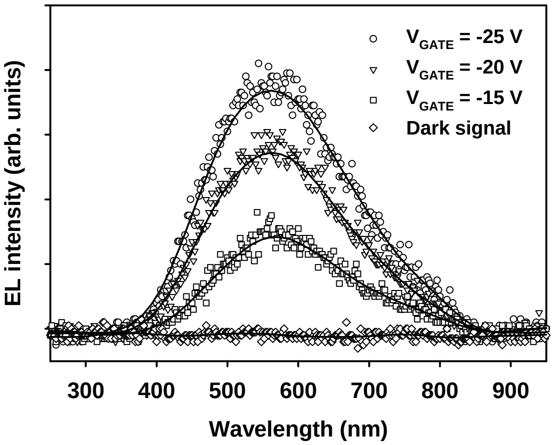

Figure 2. Typical EL spectra obtained from the nc-Al/AlN nanocomposite thin film under different V_{GATE}.

With the application of a negative gate voltage (V_{GATE}) to the ITO gate electrode, visible EL with yellow color can be obtained from the nc-Al/AlN nanocomposite thin film. As shown in Figure 2, a broad EL peak centered at ~ 565 nm (2.19 eV) can be observed, and the EL intensity increases with the magnitude of V_{GATE}. Note that when the polarity of V_{GATE} is positive, no EL can be produced. The EL cannot be generated from the ITO gate electrode, because the light emission from the gate electrode usually happens as a result of the impact of hot electrons to the gate electrode biased with a positive gate voltage [4]. In the present case, the ITO electrode is negatively biased to supply electrons during the EL process. Therefore, the visible EL is from the nc-Al/AlN thin film.

The current transport behavior in the nc-Al/AlN thin film was studied by the current-voltage (J_{GATE}-V_{GATE}) measurements in order to understand the EL behaviors. From the room-temperature J_{GATE}-V_{GATE} curve shown in Figure 3, a power-law relationship between J_{GATE} and V_{GATE} can be observed. On the other hand, the integrated EL intensity as a function of the magnitude of the V_{GATE} is also shown in Figure 3. The dependence of the EL intensity on the V_{GATE} also shows a similar trend as that of the current conduction. The results indicate that the light emission is directly related to the carrier transport in the thin film.

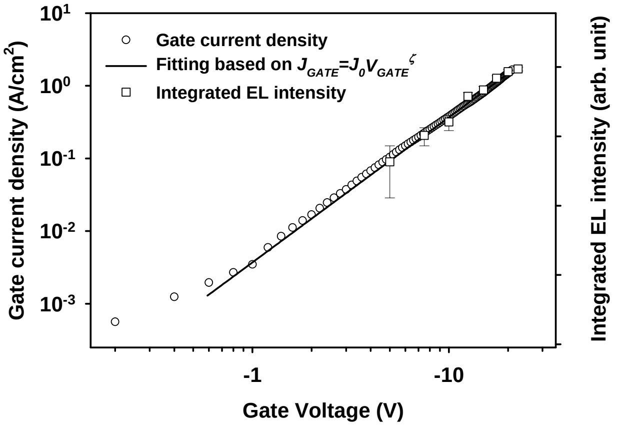

Figure 3. Power-law dependence of the gate current density (J_{GATE}) and the integrated EL intensity on the gate voltage (V_{GATE}) at room temperature.

The current transport in the nc-Al/AlN thin film can be explained by the percolation concept [3]. As shown in Figure 4(a), many tunneling paths are formed in the thin film by the nc-Al arrays, where the nanocrystals act as tunneling sites for the injected electrons. Due to the existence of many nanocrystals randomly distributed in the thin film, percolation networks of the tunneling paths which electrically connect the ITO gate to the Si substrate are formed. This greatly enhances the current conduction in the thin film, which plays an important role in the EL process.

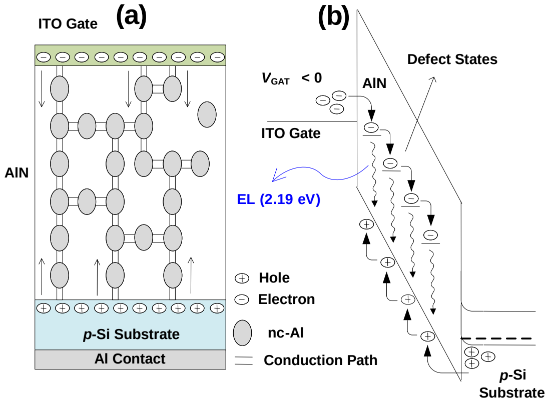

Figure 4. (a) Schematic illustration of the carrier transport along the conduction paths; (b) The radiative recombination of the transported electrons and holes occurs via the deep-level defects along the nc-Al tunneling paths.

Under the application of a negative V_{GATE}, holes from the *p*-Si substrate are injected into the nc-Al/AlN thin film and they are transported via the tunneling paths. In the mean time, electrons from the ITO gate are also injected into the thin film and they also travel along the tunneling paths. Along the tunneling paths, the radiative recombination of the injected electrons and holes could occur and give rise to the yellow EL. The light emission mechanism is illustrated in Figure 4(b). It is known that deep-level defects with energy levels ~ 3.4 – 4.5 eV below the conduction band edge of AlN exist in the AlN thin film [5]. When the injected electrons are transported along the tunneling paths formed by nc-Al, some of the electrons are trapped by the deep-level defects. The radiative recombination of the trapped electrons with the holes in the valence band of AlN injected from the *p*-Si substrate produces light emission with energies of ~ 1.7 – 2.8 eV (the bandgap for AlN is ~ 6.2 eV). That explains the observed EL peak centered at 2.19 eV.

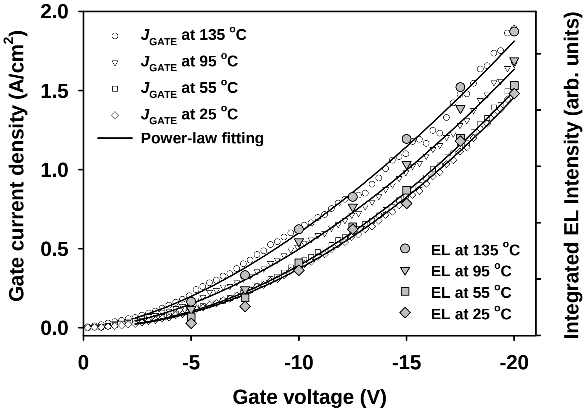

Figure 5. Integrated EL intensity and current transport as functions of gate voltage at various temperatures.

The EL and current transport behavior were also studied at elevated temperatures ranging from 55 ºC to 135 ºC. EL spectra which are similar to those shown in Figure 3 are observed under elevated temperatures, and the dependence of both the current and the integrated EL intensity on the V_{GATE} is also similar to that of the room-temperature measurement. As shown in Figure 5, both the current transport and the integrated EL intensity follow a power law at the elevated temperatures. As the temperature increases, both the current conduction and the integrated EL intensity are enhanced, suggesting that the enhancement in EL intensity at elevated temperature is due to the enhanced current conduction. The fact that the current conduction exhibits a moderate enhancement at elevated temperature as shown in Figure 5 suggests that the charge transfer via the tunneling paths formed by the nc-Al could be a thermally-assisted tunneling process.

The endurance behavior of the light emitting device was measured by applying a sequence of pulse voltage to the ITO gate electrode to turn on and off the device. During the "on" state, the magnitude of the pulse voltage is -20 V and the width of the pulse voltage is 10 ms. For the "off" state, the applied voltage is kept at 0 V. The EL intensity was recorded during the endurance measurement. The measurement was done at four different temperatures ranging from 25 ºC (room-temperature) to 135 ºC. As shown in Figure 6, no obvious decay in the EL intensity is observed during the test of up to 10^6 on/off cycles. The result indicates the excellent light emission endurance of the nc-Al/AlN nanocomposite thin film.

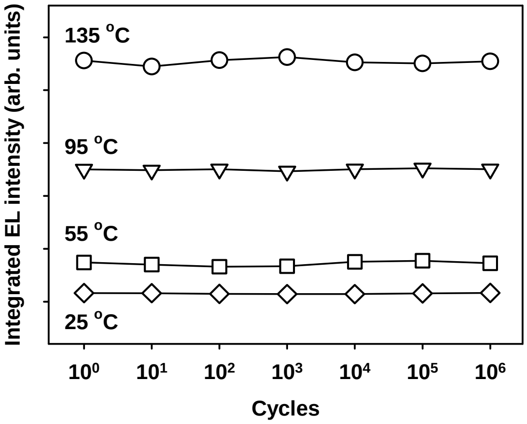

Figure 6. Endurance characteristics of the light emission at various temperatures. The pulsed gate voltage is -20 V.

4 CONCLUSION

Yellow EL with a spectrum peaked at 565 nm from sputtering synthesized AlN nanocomposite thin film containing nc-Al has been observed. The EL intensity is linearly related to the current transport in the thin film, and both the current transport and the EL intensity exhibit a power-law dependence on the applied voltage. The formation of percolation networks by nc-Al in the AlN matrix is used to explain the current transport, and the radiative recombination of the injected electrons and holes via the deep-level defects is used to explain the EL. Measurements done at elevated temperature shows that the EL is enhanced due to the enhanced current transport at higher temperature. Excellent endurance has been observed. The results suggest the potential applications of the nc-AlN/AlN thin film in the low-cost and CMOS-compatible optoelectronic devices. This work has been financially supported by the National Research Foundation of Singapore under project No. NRF-G-CRP 2007-01.

REFERENCES

[1] E. S. Dettmer, B. M. Romenesko, H. K. Charles, Jr., B. Carkhuff, and D. J. Merrill, IEEE Trans. Compon., Hybrids, Manuf. Technol. 12, 543, 1989.
[2] Y. Liu, T. P. Chen, P. Zhao, S. Zhang, S. Fung, and Y. Q. Fu, Appl. Phys. Lett. 87, 033112, 2005.
[3] Y. Liu T. P. Chen, H. W. Lau, J. I. Wong, L. Ding, S. Zhang, and S. Fung, Appl. Phys. Lett. 89, 123101, 2006.
[4] D. J. DiMaria, J. R. Kirtley, E. J. Pakulis, D. W. Dong, T. S. Kuan, F. L. Pesavento, T. N. Theis, J. A. Cutro, and S. D. Brorson, J. Appl. Phys. 56, 401, 1984.
[5] T. L. Tansley and R. J. Egan, Phys. Rev. B 45, 10942, 1992.