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Room-Temperature Visible Electroluminescence From Aluminum Nitride Thin Film Embedded With Aluminum Nanocrystals

Ming Yang, *Student Member, IEEE*, T. P. Chen, Yang Liu, Liang Ding, Jen It Wong, *Student Member, IEEE*, Zhen Liu, Sam Zhang, Wali Zhang, and Furong Zhu

***Abstract*—In this brief, room-temperature visible electroluminescence (EL) from aluminum nitride (AlN) thin films containing aluminum nanocrystals (nc-Al) prepared by a radio-frequency magnetron sputtering technique is reported. The EL shows a broad spectrum peaked at 565 nm (2.19 eV) when a negative gate voltage is applied. A linear relationship between the EL and the current transport in the nc-Al/AlN thin film system is observed, and both the current transport and the EL intensity exhibit a power-law dependence on the gate voltage. These results are explained in terms of the formation of percolation networks of tunneling paths by the nc-Al arrays and the radiative recombination of the injected electrons and holes via the deep-level defects at the locations of nc-Al along the tunneling paths.**

***Index Terms*—Aluminum nanocrystal, aluminum nitride, electroluminescence, light emitting device.**

I. Introduction

Aluminum nitride (AlN) has promising applications in surface acoustic wave [1], [2], memory [3], [4], and optoelectronic devices [5]–[8]. AlN exhibits outstanding physical properties, such as a wide bandgap (6.2 eV) [9], high thermal conductivity (320 W/mK) [10], and good match of both thermal expansion coefficient and lattice constant to those of Si substrates [10], [11]. A number of techniques can be used to fabricate AlN films, including reactive evaporation [12], chemical vapor deposition [13], molecular beam epitaxy [14], pulsed laser deposition [15], and reactive magnetron sputtering [1], [2], [7]. Among them, the sputtering technique is widely used due to its advantages of fast growth rate and low cost. Recently, amorphous AlN thin film containing aluminum nanocrystals (nc-Al) has been successfully deposited on Si substrates by a radio-frequency (RF) magnetron sputtering technique to form a nanocomposite thin film. It has been demonstrated that such nc-Al/AlN nanocomposite thin film possesses memory effect [4] and interesting current conduction behaviors [16]. The conduction modulation caused by the charge trapping and detrapping in such nc-Al/AlN thin film under ultraviolet (UV) illumination has also been observed [17]. In this brief, we report visible electroluminescence (EL) from the nc-Al/AlN thin films at room temperature.

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II. Experimental Details

The nc-Al/AlN thin film was deposited on p-type ⟨100⟩-oriented Si substrate by the technique of RF magnetron sputtering. The purity of the Al target used in the sputtering is 99.999%. The sputtering was performed in a gaseous mixture of Ar and N_2. Prior to the deposition, both the Al target and the Si substrate were cleaned by the sputtering. The base pressure of the chamber was lower than 1.0×10^{-7} torr. During the deposition, the flow rates for Ar and N_2 were 15 and 45 sccm, respectively. The RF power during deposition was 500 W, and the substrate temperature was 400 °C. The thickness of the thin film deposited on the Si substrate is ~34 nm. An identical thin film deposition was also carried out on a fused silica substrate for the absorption study. To form the metal–insulator–semiconductor (MIS) light-emitting structure, the wafer backside was coated with an aluminum layer with the thickness of 200 nm to form the backside contact. Subsequently, the array of indium tin oxide (ITO) gate electrodes, with a diameter of 2.5 mm for each electrode, was formed on the sample surface.

X-ray photoemission spectroscopy (XPS) and cross-sectional transmission electron microscopy (TEM) were used to investigate the chemical and structural properties of the nc-Al/AlN thin films, respectively. Ar ion etching of the film surface (about 5 nm in depth) was performed prior to the XPS measurement to remove the Al oxide present in the surface region [4]. A Keithley 2400 semiconductor characterization system was used for both the current–voltage (I–V) and EL measurements. The light-emission measurement was carried out with a Dongwoo Optron PDS-1 photomultiplier tube (PMT) detector (in current-sensing mode) and a Dongwoo Optron DM150i monochromator (the wavelength range is 185–1600 nm; the resolution is 0.2 nm). To obtain the

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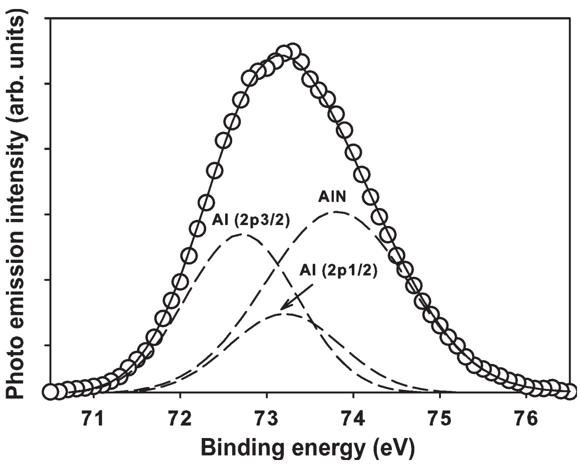

Fig. 1. Gaussian peak deconvolution of the Al $2p$ core level from the XPS measurement of the nc-Al/AlN thin film.

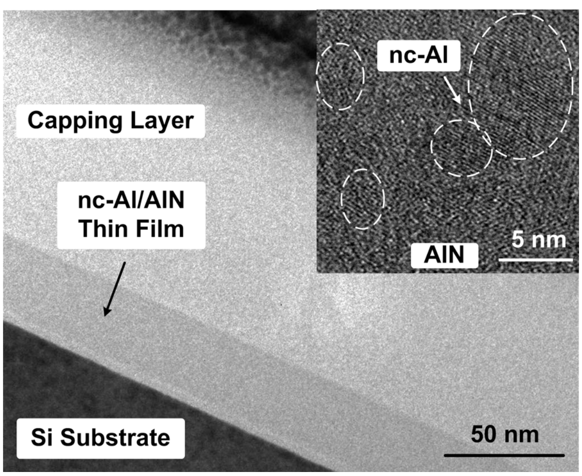

Fig. 2. Cross-sectional TEM image of the nc-Al/AlN thin film. The inset shows the existence of nc-Al embedded in the AlN matrix.

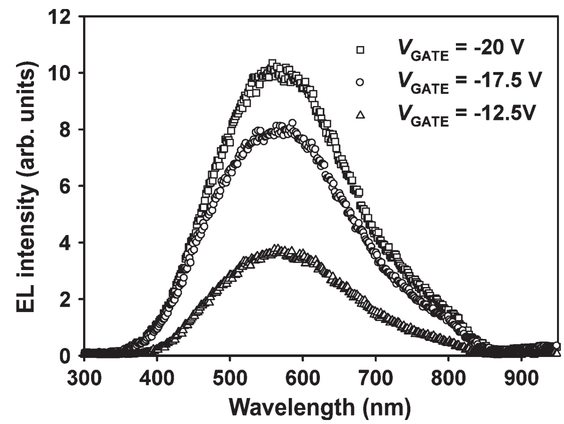

Fig. 3. EL spectra obtained from the nc-Al/AlN thin film under different V_{GATE}.

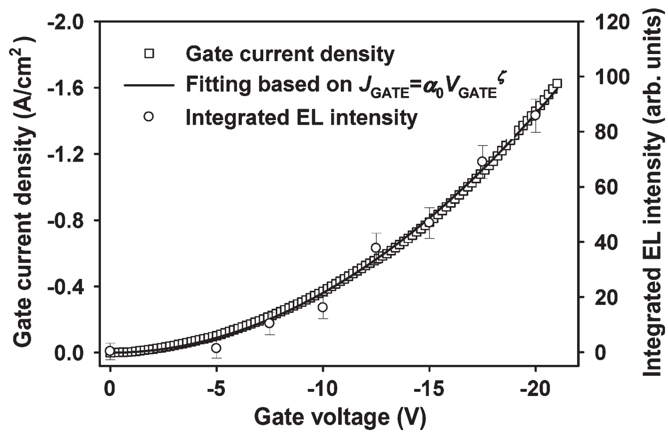

Fig. 4. Power-law dependence of the gate current density (J_{GATE}) and the integrated EL intensity on the V_{GATE}.

absorption coefficient of the thin films, transmission and reflection measurements were carried out with a Shimadzu UV-2450 spectrometer in the wavelength range of 190–850 nm. All the measurements were conducted in a dark environment at room temperature.

III. Results and Discussions

Fig. 1 shows the peak deconvolution of the Al $2p$ core level obtained from the XPS measurement. As shown in the figure, the XPS Al $2p$ core level can be decomposed into three Gaussian peaks, namely the AlN, Al $2p1/2$, and Al $2p3/2$ peaks. As an Ar ion etching of the film surface (about 5 nm in depth) was performed prior to the XPS measurement to remove the Al oxide present in the surface region [4], no significant Al_2O_3 signal was detected in this brief. The area ratio of the AlN peak to the Al peaks confirms that the deposited thin film is Al-rich. The cross-sectional TEM image for the nc-Al/AlN thin film is shown in Fig. 2. The nc-Al with lattice fringes embedded in the amorphous AlN matrix can be observed from the high-resolution TEM image shown in the inset of Fig. 2.

The nc-Al/AlN thin film exhibits a visible EL with yellow color when a negative gate voltage (V_{GATE}) is applied to the ITO gate electrode. Fig. 3 shows the typical EL spectra for different V_{GATE}'s. A broad but prominent EL band extending from 350 to 850 nm can be clearly observed, and the EL intensity becomes stronger as the magnitude of the negative V_{GATE} increases. The EL peak is located at ~565 nm (2.19 eV). When the polarity of V_{GATE} is positive, no EL can be induced regardless of the magnitude of the V_{GATE}. The EL cannot be due to the ITO gate electrode itself, considering that 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 [18]. In our case, the ITO electrode is negatively biased to supply electrons during the EL measurement. Therefore, the nc-Al/AlN thin film is responsible for the visible EL.

The current transport behavior in the nc-Al/AlN thin film is studied with the I–V measurement in order to understand the EL behaviors. Fig. 4 shows the gate current density (J_{GATE}) as a function of the magnitude of the V_{GATE}. The curve fitting suggests that the J_{GATE} and the V_{GATE} have a power-law relationship

$$J_{\mathrm{GATE}} = \alpha_0 V_{\mathrm{GATE}}^{\zeta} \quad (1)$$

where α_0 is a coefficient and ζ is the scaling exponent. From the curve fitting, it is found that the scaling exponent $\zeta \approx 2$.

Note that ζ is affected by the concentration and distribution of nanocrystals, as well as the charge trapping in the nanocrystals [16], [17]. In particular, ζ could be changed by the application of a voltage or even an I–V measurement itself due to the change in the charging state [16]. The power-law behavior of current transport has been reported for arrays of small metallic dots [19] and metal nanocrystal arrays [20]. The current transport in the nc-Al/AlN thin film can be explained by the percolation concept [16]. 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.

The integrated EL intensity as a function of the magnitude of the V_{GATE} is also shown in Fig. 4. As can be observed from the figure, the dependence of the EL intensity on the V_{GATE} also follows a power law which has the same trend as that of the current transport, showing a linear relationship between the current transport and the EL intensity. This indicates that the light emission is directly related to the carrier transport in the thin film rather in the ITO gate or at the interface between the ITO gate (or the p-Si substrate) and the thin film. It also implies that the radiative recombination of the injected electrons and holes could occur along the tunneling paths formed by the nc-Al, regardless of the actual recombination processes.

The radiative recombination of the electrons and holes could be associated with defects in the nc-Al/AlN thin film. Various defects, including Al vacancy (V_{Al}), N vacancy (V_{N}), and Al and N antisite defects ($\mathrm{Al_N}$ and $\mathrm{N_{AL}}$), exist in the AlN system [21], [22]. The material system in this brief is actually an Al-rich AlN thin film with the existence of a large amount of nc-Al. As revealed by the TEM images, the interfaces between the nc-Al and the AlN are imperfect. Due to the imperfection of the AlN matrix with the embedded nc-Al, it is reasonable to assume that there are large amounts of defects at the interfacial regions between the embedded nc-Al and the AlN matrix. Aside from the aforementioned defects, the oxygen impurity may also contribute to the luminescence due to the deep-level defects created by the substitution of nitrogen in the AlN with oxygen [23], [24]. However, the oxygen-related defects are unlikely to be the major contributors to the EL in this brief, as no significant signal of oxygen impurity was detected in the XPS measurement on our samples. Nevertheless, the absorption spectrum of the nc-Al/AlN thin film shown in Fig. 5 suggests the existence of defects in the thin film. As can be seen in the figure, the nc-Al/AlN thin film shows a band-to-band absorption edge (~6.2 eV) similar to that of the pure AlN [25] and a very broad band tail. The broad band tail is indicative of a number of possible electronic defect structures present in the thin film [23].

As pointed out earlier, both the EL intensity and the current transport follow a similar power-law dependence on the gate voltage, which suggests that the radiative recombination of the transported electrons and holes occurs at the locations of nc-Al along the tunneling paths. Therefore, if the defects are responsible for the EL, then the defects at the interfaces between nc-Al and AlN, rather than those in the amorphous AlN matrix, play the major role. It is worth mentioning that the existence of embedded nc-Al in the AlN matrix has two roles; one is to enhance the current transport by forming a large number of conductive tunneling paths, and the other is to introduce the interfacial luminescence defects along the tunneling paths. The radiative recombination of the injected electrons and holes in the nc-Al/AlN thin film is thus associated with these defects at the locations of nc-Al. Fig. 6 shows a possible defect-related radiative recombination process for the observed EL in the nc-Al/AlN thin film. The defects involved in the process are those deep-level defects with energy levels ~3.4–4.5 eV below the conduction band edge of AlN [21]. As mentioned previously, under a negative V_{GATE}, electrons and holes are injected from the ITO gate and the p-type Si substrate, respectively. 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 (note that the AlN bandgap is ~6.2 eV). This

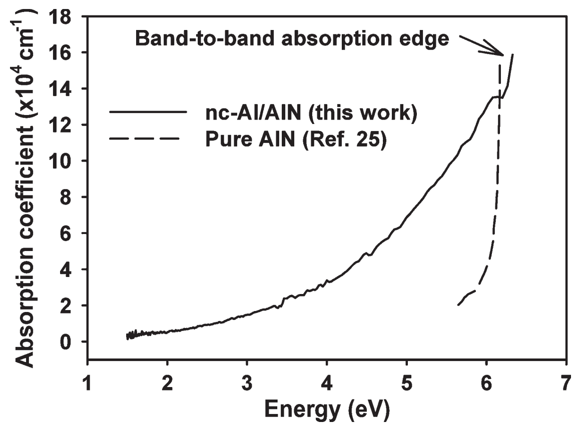

Fig. 5. Room-temperature absorption spectrum of the nc-Al/AlN thin film. The corresponding spectrum of pure AlN (from [25]) is also presented for comparison.

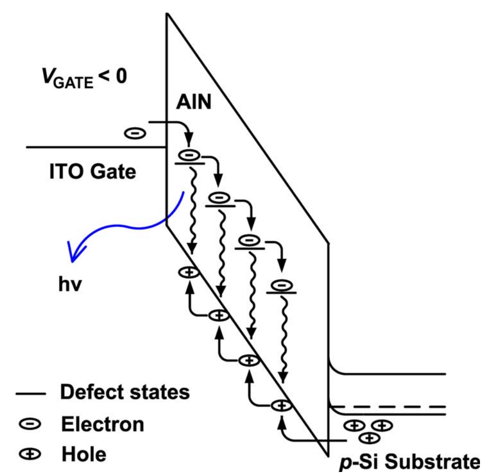

Fig. 6. Schematic energy band diagram of the light-emitting structure and a possible radiative recombination process of the injected electrons and holes via the deep-level defects at the location of nc-Al along the nc-Al tunneling paths.

explains the observed EL peak centered at 2.19 eV. It also explains why the current transport and the EL intensity exhibit a similar power-law dependence on the V_{GATE}.

IV. Conclusion

In summary, room-temperature visible EL with a broad spectrum peaked at 565 nm (2.19 eV) from AlN thin film containing nc-Al fabricated by RF magnetron sputtering technique has been observed. It is found that the EL intensity is linearly related to the current transport in the nanocomposite system, and both the current transport and the EL intensity exhibit a power-law dependence on the applied voltage. The current conduction is explained in terms of the formation of percolation networks of the tunneling paths by nc-Al in the AlN matrix. 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.

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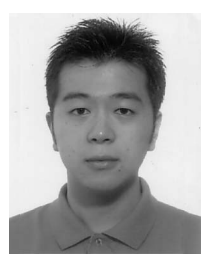

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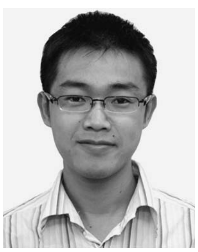

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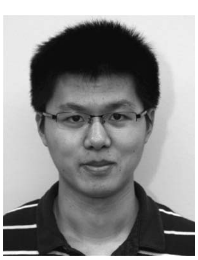

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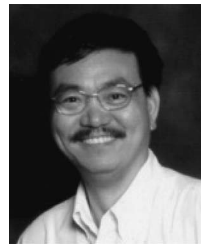

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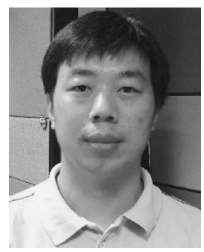

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