Green electroluminescence from a Tb-doped AlN thin-film device on Si

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Green photoluminescence and electroluminescence (EL) from Tb implanted AlN films have been observed at room temperature. The AlN films of 180 nm thickness were grown on *n*-type Si(111) by metalorganic chemical vapor deposition. X-ray diffraction shows that the AlN is polycrystalline. The AlN films were doped by ion implantation of Tb to a peak concentration of 1 at. %. A postimplantation annealing step was required to obtain optically active Tb ions. A dc EL device was fabricated using a transparent ZnO:Al top electrode. The strong room-temperature green light emission was observable with the naked eye. It was obtained with a drive current density of 2–70 mA/cm^2 at a drive voltage of 70–100 V. The emission lines between 490 and 650 nm originate from Tb^{3+} transitions from the $^5D_4$ level to ground state multiplets. The observed luminescence lifetimes are approximately 0.5 ms. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1497461]

I. INTRODUCTION

Silicon-based optoelectronics are still in need of an efficient optical emitter. Rare-earth (RE) doped crystalline Si has been used as a host for IR emission. Since high-quality crystalline Si detectors are available for visible wavelength, a compatible Si-based light source is highly desirable. Wide band gap hosts as SiO_2, Si_3N_4, SiC, GaN, or AlN may be deposited on Si and used as hosts for ions, which emit visible luminescence. Electrical excitation of these ions is possible, although, at present, it is not as efficient as the band to band electroluminescence (EL) of direct band gap semiconductors. RE doped wide band gap materials emit at individual characteristic wavelengths and allow different excitation processes, especially electron–hole recombination at the RE dopants or hot carrier impact excitation of the RE ions. Among the III–V semiconductors, recently, GaN has been studied very successfully.[1–4] In addition to its excellent chemical stability, the wide band gap of 3.4 eV makes GaN a suitable host for RE ions, permitting the optical emission of a wide spectrum of wavelengths. In comparison, AlN has received much less attention, although it is successfully in use as a buffer layer for the growth of GaN. Undoped AlN is highly resistive ($\rho = 10^7 - 10^{13}\ \Omega$ cm) and intentional doping appears to be difficult. The doping issues of AlN have been carefully analyzed and compared to the situation in GaN and AlGaN alloys.[5] However, AlN has some outstanding physical properties, such as hardness, high thermal conductivity, a high melting temperature, and a reasonable match of the thermal expansion coefficient to a Si substrate.[6] Its wide band gap of 6.2 eV indicates that it is suited for RE-based optical emission. This is not only due to the large wavelength range but also due to the lower thermal quenching of the RE luminescence, which is inversely proportional to the band gap of the host semiconductor.[7,8] Cathodoluminescence and photoluminescence (PL) of RE-doped AlN have been reported by several groups.[9–11] Visible EL emission from a RE-doped AlN film was observed from an Er-doped alternating-current thin-film EL structure.[12] In this article, we present Tb-implanted AlN dc EL device.

II. EXPERIMENTS AND RESULTS

Thin films of AlN were grown on 2 in. diameter *n*-type Si(111) substrates by metalorganic chemical vapor deposition (MOCVD) at 720 °C. The thickness of films was 185 nm. This was confirmed by Rutherford backscattering spectrometry (RBS). RBS channeling and x-ray diffraction revealed that all films were polycrystalline. In order to study the PL of Tb ions in AlN, Tb ions were implanted at room temperature. The fluence was 2.7×10^{15} Tb/cm^2 at 200 keV and 5.4×10^{15} Tb/cm^2 at 400 keV, leading to a maximum Tb concentration of 1 at. %, located in the center of the insulating film. A previous PL study of the concentration dependence of Tb in SiO_2 had shown that a higher Tb concentration leads to strong crossrelaxation.[13] The computer code TRIM was used to determine the implantation parameters. The resulting Tb depth profile was verified by RBS and is shown in Fig. 1. To optimize the luminescence intensity of AlN:Tb, the samples were annealed for 30 min at 750 °C, 850 °C, 950 °C, or 1050 °C in a nitrogen ambient. Annealing at any of these temperatures did not result in a noticeable diffusion of the RE dopants. One example is given in Fig. 1. An Ar

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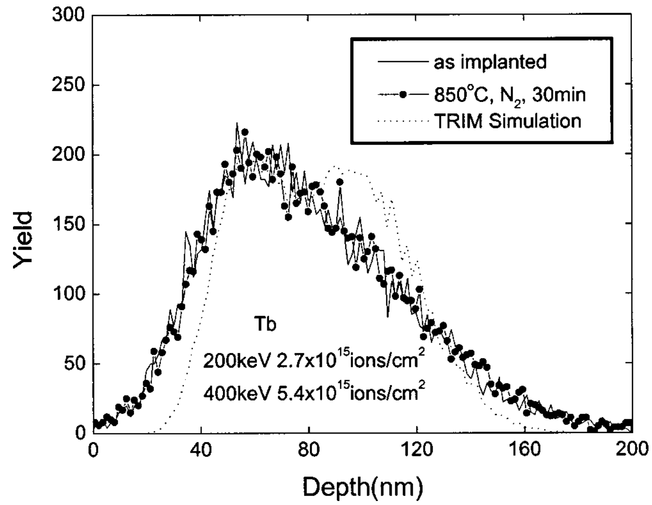

FIG. 1. RBS spectrum, showing the depth profile of implanted Tb in AlN before and after annealing.

laser ($\lambda = 351$ nm) was used to excite the PL. This energy matches the intra-4f transition from the ground level $^7F_6$ to the excited $^5L_9$ level.[13,14] The measured spectra are shown in Fig. 2. Annealing at 850 °C gave the strongest integrated PL intensity.

Based on these findings, the devices for EL were fabricated. For a schematic drawing, see Fig. 3. First, a layer of 200 nm SiO_2 was deposited onto the as-grown AlN by plasma enhanced chemical vapor deposition in order to prevent any contamination of the AlN surface and to provide the overall electrical insulation. As the next step, windows were etched into the SiO_2. The diameter of the windows varied from 0.2 to 1 mm. Then, the Tb was implanted as mentioned herein, giving again a maximum concentration of 1 at. %. The samples were annealed at 850 °C in N_2 for 30 min.

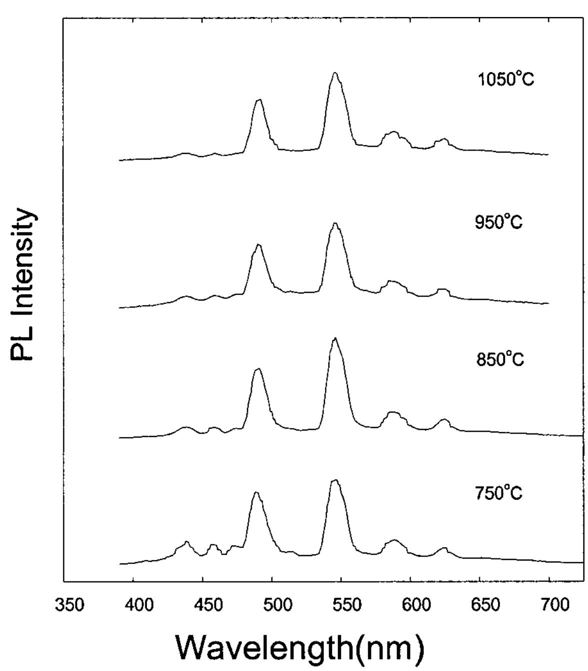

FIG. 2. PL spectra of Tb implanted AlN after annealing at different temperatures.

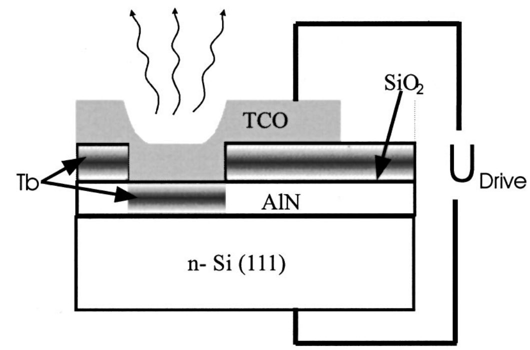

FIG. 3. Schematic drawing of the AlN:Tb EL device. The Tb in the SiO_2 is not pumped by the drive current and is not active in the EL experiments.

Finally, a transparent top electrode was deposited. We used 300 nm of ZnO:Al. This electrode has a transmission of approximately 80%–85% for green light. The bottom contact was formed by the Si substrate. Similar devices were fabricated to study SiO_2 as the host for the RE ions. In this case, the AlN layer was replaced by another SiO_2 layer. The EL signals were collected using a spectrometer, a photomultiplier, and a lock-in amplifier. All of the measurements were made at room temperature.

Figure 4 shows the current–voltage (I–V) characteristics of a 0.25 mm^2 structure. The band alignment of the heterostructure seems fairly symmetrical, since the current is essentially independent of the sign of the applied voltage. "Forward" denotes a positive voltage applied to the ZnO top electrode. Tunneling sets in at about 30 V, which corresponds to a field of about 1.6 MV/cm across the AlN. At higher fields, an exponential increase is observed. These I–V characteristics of AlN are similar to those of SiO_2 although no tunneling current was observed in SiO_2 at fields below 6 MV/cm.[15] EL was excited by current pulses of 22 ms duration and 20 Hz repetition. The visible part of the EL spectrum is shown in Fig. 5. The green light was observable with the naked eye. The lines at 492, 548, 591, and 625 nm correspond to the transitions from the $^5D_4$ level to the ground state multiplets ^7D_J. The transition from $^5D_4$ to $^7F_5$ of Tb dominates the green emission. No blue transitions from the $^5D_3$ level were seen. This may be due to the Tb concentration of 1%. At this concentration, the crossrelaxation between two excited $^5D_4$ and $^5D_3$ states to $^7F_6$ and $^7F_0$ ground

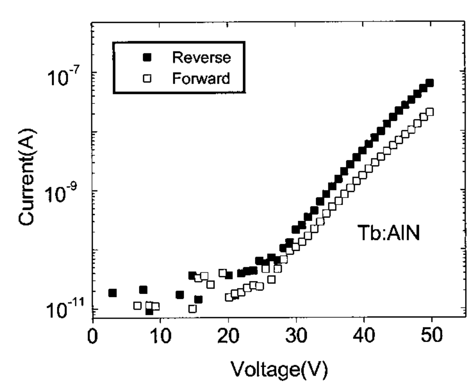

FIG. 4. I–V characteristics of the AlN:Tb structure shown in Fig. 3.

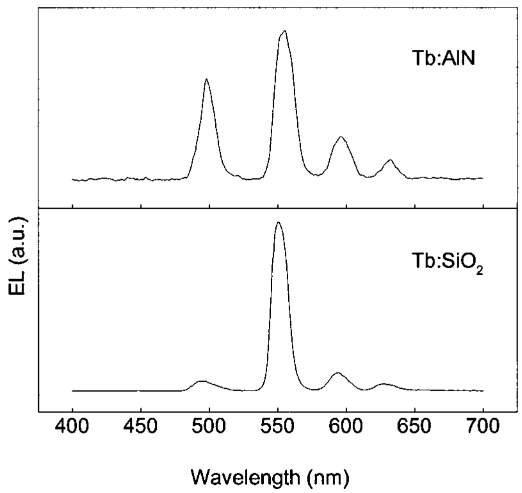

FIG. 5. EL spectra of AlN:Tb and SiO_2:Tb.

states sets in.[12] The observed linewidths (full width at half maximum) of the $^5D_4$ to $^7F_6$ and $^5D_4$ to $^7F_5$ transitions are 12 and 17 nm, respectively. These are the same linewidths as observed in the well-known spectra emitted from Tb within a host of ZnS.[16] Figure 6 shows the time dependence of the EL signal at $\lambda=548$ nm, recorded by an oscilloscope. The decay time constant $\tau\approx0.5$ ms is similar to the Tb luminescence lifetime in other semiconductors.[10,13,17] The intensity of the emission at $\lambda=548$ nm is plotted as a function of bias current in Fig. 7. The EL intensity increases stronger than linear with the bias current. For low current densities, we find $EL\sim j^{1.8}$, at current densities exceeding 35 mA/cm^2 the luminescence intensity scales as $EL\sim j^{3.2}$. A stronger than linear increase is expected, because at higher electrical fields not only the number of hot carriers increases, but also their average kinetic energy. At higher carrier energies, increasing excitation cross sections are observed, since more Tb states (higher excitation energy) are involved. No saturation is observed even at a current density as high as 70 mA/cm^2, which is the maximum current density used. The green EL emission spectra from Tb:AlN and Tb:SiO_2 are similar, but for the same bias current, the EL intensity from Tb:AlN is only one third of that from Tb:SiO_2. The external EL quantum efficiency of the Tb:SiO_2 device was 4×10^{-5} (Ref. 18) and the external

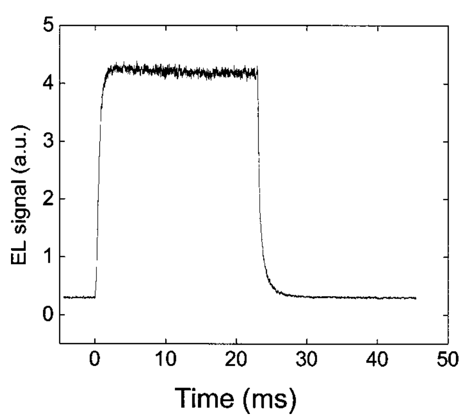

FIG. 6. EL signal from AlN:Tb, recorded by an oscilloscope.

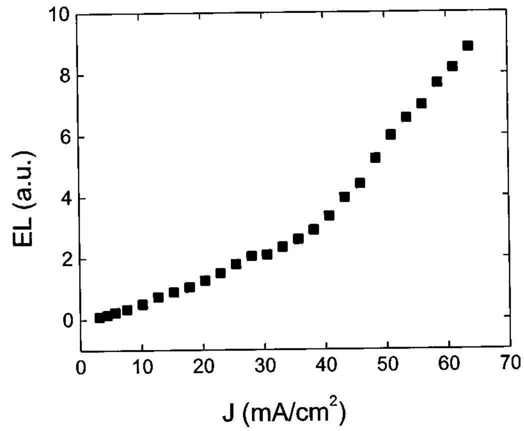

FIG. 7. EL intensity from AlN:Tb vs drive current.

quantum efficiency of Tb:AlN is estimated to be 1.5×10^{-5}, considering the Fresnel reflections (optical index of AlN: $n=2.2$) and the photon emission cone. Note, that the internal quantum efficiency of this structure is strongly dependent on the quality and homogeneity of the AlN film, since any low resistance current path will not contribute to the EL excitation and deteriorate the device efficiency.

III. SUMMARY

In summary, we have reported green emission from a Tb implanted AlN EL device. Polycrystalline AlN films were prepared by MOCVD on Si(111). 1 at. % of Tb was introduced by ion implantation. After annealing at a moderate temperature, the PL and EL emission spectra of Tb:AlN show four lines in the visible range, which correspond to the transitions from $^5D_4$ to ground state multiplets. As a polycrystalline film of AlN on Si is fairly easy to grow and dope by ion implantation, the presented system has significant potential for applications.

ACKNOWLEDGMENT

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[1] R. Birkhahn, M. Garter, and A. J. Steckl, Appl. Phys. Lett. **74**, 2161 (1999).
[2] J. Heikenfeld, M. Garter, D. S. Lee, R. Birkhahn, and A. J. Steckl, Appl. Phys. Lett. **75**, 1189 (1999).
[3] A. J. Steckl, M. Garter, D. S. Lee, J. Heikenfeld, and R. Birkhahn, Appl. Phys. Lett. **75**, 2184 (1999).
[4] D. S. Lee, J. Heikenfeld, R. Birkhahn, M. Garter, B. K. Lee, and A. J. Steckl, Appl. Phys. Lett. **76**, 1525 (2000).
[5] A. Fara, F. Bernardini, and V. Fiorentini, J. Appl. Phys. **85**, 2001 (1999).
[6] *Properties of Group III Nitrides*, edited by J. H. Edgar (emis data reviews No. 11, IEE-INSPEC, London, 1994).
[7] P. Favennec, H. L'Haridon, M. Salvi, D. Moutonnet, and Y. Le Guillou, Electron. Lett. **25**, 718 (1989).
[8] S. J. Pearton, C. R. Abernathy, J. D. Mackenzie, U. Hömmerich, J. M. Zavada, R. G. Wilson, and R. N. Schwartz, J. Vac. Sci. Technol. A **16**, 1627 (1998).
[9] K. Gurumurugan, H. Chen, G. R. Harp, W. M. Jadwisienczak, and H. J. Lozykowski, Appl. Phys. Lett. **74**, 3008 (1999).
[10] W. M. Jadwisienczak, H. J. Lozykowski, F. Perjeru, H. Chen, G. R. Harp, M. Kordesch, and I. G. Brown, Appl. Phys. Lett. **76**, 3373 (2000).
[11] M. L. Caldwell, A. L. Martin, V. I. Dimitrova, P. G. Van Patten, M. E.

Kordesch, and H. H. Richardson, Appl. Phys. Lett. **78**, 1246 (2001).
[12] V. I. Dimitrova, P. G. Van Patten, H. H. Richardson, and M. E. Kordesch, Appl. Phys. Lett. **77**, 478 (2000).
[13] H. Amekura, A. Eckau, R. Carius, and C. Buchal, J. Appl. Phys. **84**, 3867 (1998).
[14] R. Reisfeld and C.K. Jorgensen, *Lasers and Excited States of Rare Earths* (Springer, Berlin, 1977), p. 140.
[15] S. Wang, A. Eckau, E. Neufeld, R. Carius, and C. Buchal, Appl. Phys. Lett. **71**, 2824 (1997).
[16] D. C. Krupka, J. Appl. Phys. **43**, 476 (1972).
[17] H. J. Lozykowski, W. Jadwisienczak, and I. Brown, Appl. Phys. Lett. **76**, 861 (2000).
[18] S. Wang, S. Coffa, R. Carius, and C. Buchal, Mater. Sci. Eng., B **81**, 102 (2001).