行政院國家科學委員會專題研究計畫 成果報告

以奈米矽或二六族兩種新材料所製程之高效率單晶片白光二極體元件

研究成果報告(精簡版)

計 畫 類 別 ：個別型
計 畫 編 號 ：NSC 95-2221-E-002-272-
執 行 期 間 ：95 年 08 月 01 日至 96 年 10 月 31 日
執 行 單 位 ：國立臺灣大學光電工程學研究所

計畫主持人：黃建璋

計畫參與人員：碩士班研究生-兼任助理：謝旻諺、謝志豪

報 告 附 件 ：出席國際會議研究心得報告及發表論文

處 理 方 式 ：本計畫可公開查詢

中 華 民 國 96 年 11 月 28 日

行政院國家科學委員會補助專題研究計畫 ■ 成 果 報 告
□期中進度報告

以奈米矽或二六族兩種新材料所製程之高效率單晶片白光二極體元件

計畫類別：■ 個別型計畫　□ 整合型計畫
計畫編號：NSC　95－2221－E －002－272－
執行期間：　95 年　8 月　1 日至　96 年 10 月 31 日

計畫主持人：黃建璋
共同主持人：
計畫參與人員：

成果報告類型(依經費核定清單規定繳交)：■精簡報告　□完整報告

本成果報告包括以下應繳交之附件：
□赴國外出差或研習心得報告一份
□赴大陸地區出差或研習心得報告一份
■出席國際學術會議心得報告及發表之論文各一份
□國際合作研究計畫國外研究報告書一份

處理方式：除產學合作研究計畫、提升產業技術及人才培育研究計畫、列管計畫及下列情形者外，得立即公開查詢
□涉及專利或其他智慧財產權，□一年□二年後可公開查詢

執行單位：台灣大學

中　華　民　國　96　年　11　月　28　日

摘要

We develop a nearly white light emitting device by integrating blue/green emission from a GaN based LED with red emission from a porous SiO2 layer. The porous SiO2 layer was fabricated by a novel process procedure to create Si nanocrystals on top of the n-type GaN layer. Red light is generated from the metal-oxide-semiconductor (Ni/Au–SiO2 oxide-n-type GaN) structure due to the electron-hole recombination in the Si nanocrystals. The device shows a blue light emission at a low biased voltage and nearly white light emission (green and red colors) at a bias voltage between 14V and 16V. Our results show the potential of applying such an integrated structure to white light illumination.

Keywords: Light emitting diodes, SiO2 porous layer, Si-nanocrystal, white light

報告內容

前言

We develop a nearly white light emitting device by integrating blue/green emission from a GaN based LED with red emission from a porous SiO_2 layer. The porous SiO_2 layer was fabricated by a novel process procedure to create Si nanocrystals on top of the n-type GaN layer. Red light is generated from the metal-oxide-semiconductor (Ni/Au–SiO_2 oxide-n-type GaN) structure due to the electron-hole recombination in the Si nanocrystals. The device shows a blue light emission at a low biased voltage and nearly white light emission (green and red colors) at a bias voltage between 14V and 16V. Our results show the potential of applying such an integrated structure to white light illumination.

研究目的

White light LEDs (light emitting diodes) have become popular due to their applications to backlights for thin film transistor-liquid crystal displays (TFT-LCDs) and general lighting. Typically, they can be fabricated with the combination of discrete red, green and blue LEDs. The phosphor converted LED is an alternative approach for white light illumination. They can be realized by mixing blue light from GaN based LEDs with yellow light converted from the blue pumped phosphor, or by mixing green, red and blue colors converted from UV pumped phosphors. Despite the availability of choices of using phosphor species for selected wavelength conversion, there are disadvantages of phosphor converted LEDs. For example, the conversion efficiency of phosphor species is typically very low. This poor energy conversion is due to a Stokes energy loss when converting short-wavelength photons to long wavelength ones. Also, the phosphor converted LEDs do not allow for the extensive tunability, particularly in terms of spectral modulation. In light of those drawbacks, direct electron-hole recombination or direct optical pump semiconductor materials are alternate candidates for high efficient light sources.

文獻探討

Silicon nanocrystals (nc-Si) embedded in SiO_2 matrix (nc-Si/ SiO_2) is attracting a lot of interest in its efficiency and stability of light emission at room temperature. Various technologies have been proposed to fabricate nc-Si structures, such as plasma enhanced chemical vapor deposition (PECVD), electron-beam evaporation, and Si-ion implantation. Photoluminescence (PL) spectra from the blue to near-infrared region were demonstrated. Even though it is generally thought that the quantum confinement effect is one of the main mechanisms for light emission, point defects, such as from Si-ion implanted SiO_2, are also considered to be another source for light emissions. In addition, electroluminescence (EL) spectra of the metal-oxide-semiconductor (MOS) structures (where the material nc-Si/SiO_2 or Si-ion implanted SiO_2 is regarded as the oxide layer and Si substrate as the semiconductor) were also reported with turn-on voltages between 3.3V and 86V in the 700~900 nm wavelength range.

研究方法

We develop a novel integrated white light emitting device. By combining the nc-Si MOS with the GaN based LED structure, i.e., by depositing a porous SiO_2 layer on top of the GaN based LED, nearly white light emission with dual spectrum peaks at 496.4 nm and 828.6 nm are demonstrated at room temperature. The turn-on voltage of the nc-Si MOS device is as low as 13.7V and the operation voltage of the nearly white light MOS-LED device is between 14V and 16V. The MOS-LED structure shows the potential of applying such an integrated device to white light generation.

The GaN based LED sample with a PL peak at around 480nm was grown on a c-plane sapphire substrate by metal-organic chemical vapor deposition (MOCVD). The material structure is composed of a 25-nm GaN buffer layer, a 1.8-μm Si-doped n-type GaN layer, a ten-period InGaN/GaN multiple quantum well structure, and a 200-nm p-type GaN layer. The patterned SiO_2 layer was deposited on top of the GaN n-type layer by PECVD with an initial thickness of 340nm. We then wet-etched the SiO_2 layer by buffer oxide etcher (BOE, NH_4F: HF=6:1) to thin down the SiO_2 layer. In the subsequent step, the sample was dry etched by inductively coupled plasma reactive ion etching (ICP-RIE) to create porous profile on SiO_2 layer. We used Ar, $SiCl_4$ and Cl_4 as etching gases with a flow rate 20sccm, 1sccm and 15sccm, respectively. The SiO_2 layer thickness was optimized by considering both the generated output optical power from the SiO_2 layer (nc-Si) and the operating voltage. Typically, a thicker SiO_2 layer is preferred for luminescence which, however, requires a higher biased voltage. In our case, the optimum thickness of the porous SiO_2 layer is around 200nm. The atomic force microscopic (AFM) surface profile of the porous SiO_2 layer grown on top of the n-type GaN layer is shown in Fig. 1. The lateral diameter of nc-Si is approximately 500nm. In the next step, Ni/Au metal layers were evaporated as the p-type electrode and were alloyed to obtain optimized ohmic contact condition. We then coated Ti/Au directly on top of the porous SiO_2 layer as the n-type contact. The Ti/Au - SiO_2 – GaN layer combination forms a MOS structure (see Fig. 2) and red emission can be observed after a 750°C annealing in our experiment. The high temperature annealing (＞700°C) deactivates the neutral oxygen vacancies defect (O≡Si−Si≡O) in the porous SiO_2 layer, and meanwhile generates nc-Si [13]. Electron-hole recombination can occur in the porous SiO_2 layer (nc-Si).

結果與討論

The EL spectra of our samples were taken at various bias voltages (see Fig. 3(a)) at room temperature. At a low bias voltage, electrons from the n-type electrode tunnel through the porous SiO_2 layer and recombine with holes in the GaN based active layer to generate blue emission. Therefore, the blue emission is dominated at low bias voltages. The peak wavelength is around 487.6nm when biased at 14V. As we increase the applied voltage to 15V, red emission is observed as the carrier recombination in the MOS structure starts to occur. It is also noted that the relative intensity of the blue emission decreases as the applied voltage increases, which is attributed to the fact that more and more carriers are recombined in the MOS region when the bias voltage increases from 14V to 15V. The blue emission is further decreased as the applied voltage is increased to 16 V. The peak wavelength of the GaN based LED is shifted to 496.4nm due to quantum confinement stark effect. The relative intensities of wavelength 496.4 nm and 828.6 nm have become comparable at the bias voltage 16V. Fig. 3(b) shows the CIE 1931 chromaticity diagram with corresponding light emitting colors at different bias voltages. With the increase of bias voltages, the bluish color (x=0.3002, y=0.3190) at 14V turns to a nearly white color (x=0.3302, 0.3338) at 16V.

We further compare the current-voltage (I-V) characteristic of the MOS- LED structure with that of the conventional LED sample (the one without any SiO_2 layer coated). As shown in Fig. 4, the I-V curve of the conventional LED shows a diode behavior (dash line), while that of the MOS-LED structure acts like a normal LED in series with a resistor when the bias voltage is below 13.7V. Since carriers from the n-type electrode of the MOS-LED structure have to to tunnel through the thin porous SiO_2 layer, the effective current under the same bias voltage of the MOS-LED structure is smaller than that of the conventional LED structure. When the bias voltage is above 13.7V (the threshold voltage of the MOS device), the device is turned on and electrons from the n-type electrode start to recombine with holes in the nc-Si. A negative resistance is observed at bias voltages above 13.7 V, indicating the generation of red light. The negative resistance in Fig. 4 corresponds to the onset of red light emission in Fig. 3 and meanwhile indicates the decrease of blue intensity. The MOS structure breaks down (blows up) at bias voltages beyond 16V, which is due to large numbers of electrons tunneling through the porous SiO_2 layer.

We develop a novel integrated white light emitting device. By integrating the nc-Si MOS with the GaN based LED structures, i.e., by depositing a porous SiO_2 layer on top of the GaN based LED, nearly white light emission with dual spectrum peaks at 496.4 nm and 828.6nm were demonstrated at room temperature. The turn-on voltage of the nc-Si MOS device is as low as 13.7V and the operating voltage of the nearly white light MOS-LED device is between 14V and 16V. In addition, a negative resistance is observed from the I-V curve of the MOS-LED structure at a bias voltage above 13.7V. It is attributed to the fact that some carriers start to recombine in the MOS region. The relative intensity of red emission is thus increased while that of the blue emission is decreased. Our results indicate the potential of applying such an integrated structure to white light generation.

圖表

Fig. 1 The AFM surface profile of the porous SiO_2 layer
Fig. 2 Schematic illustration of a GaN based LED integrated with a porous SiO_2 layer
Fig. 3 Room-temperature EL spectra (a) and CIE 1931 chromaticity diagram (b) of the light emission of LEDs at bias voltages 14 V, 15 V and 16 V.
Fig. 4 I-V curves of the MOS-LED sample (solid line) and the conventional LED (dash line)

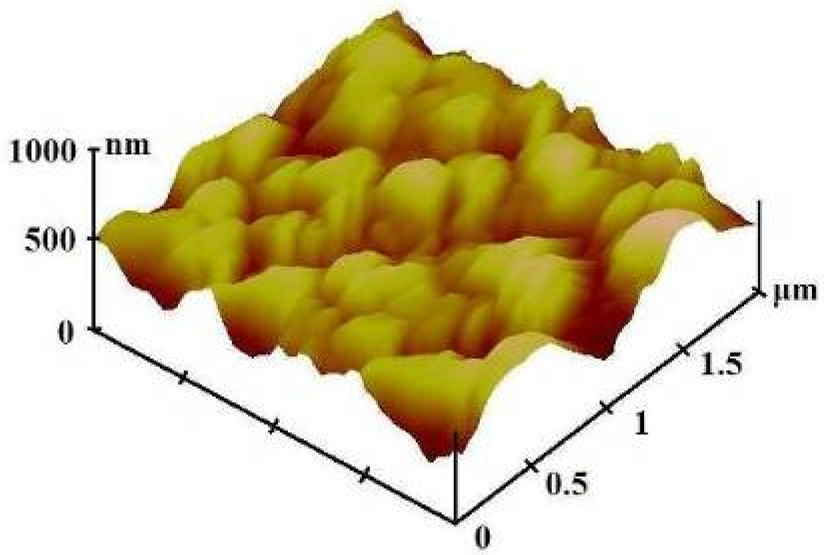


Fig. 1

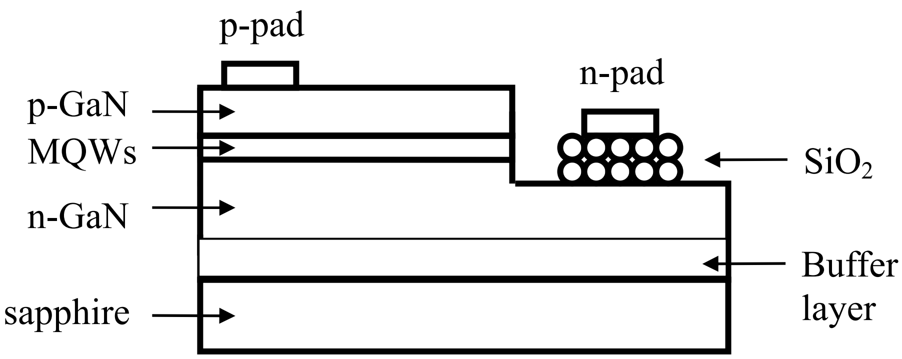
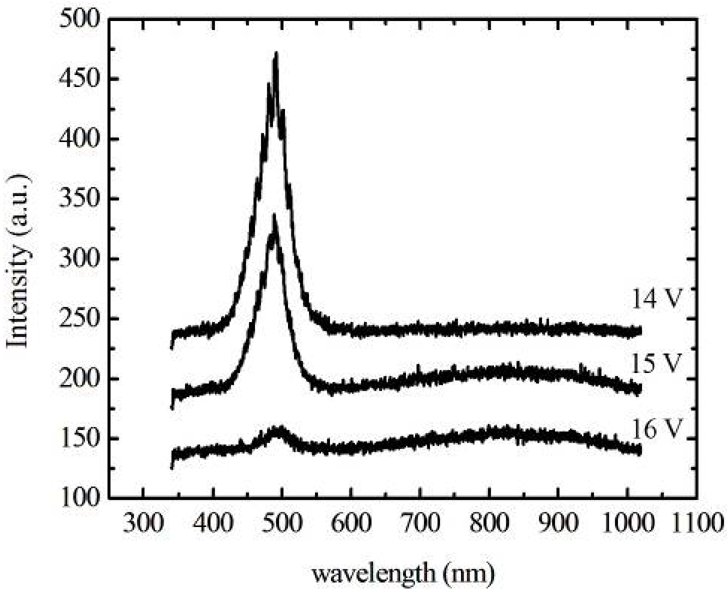


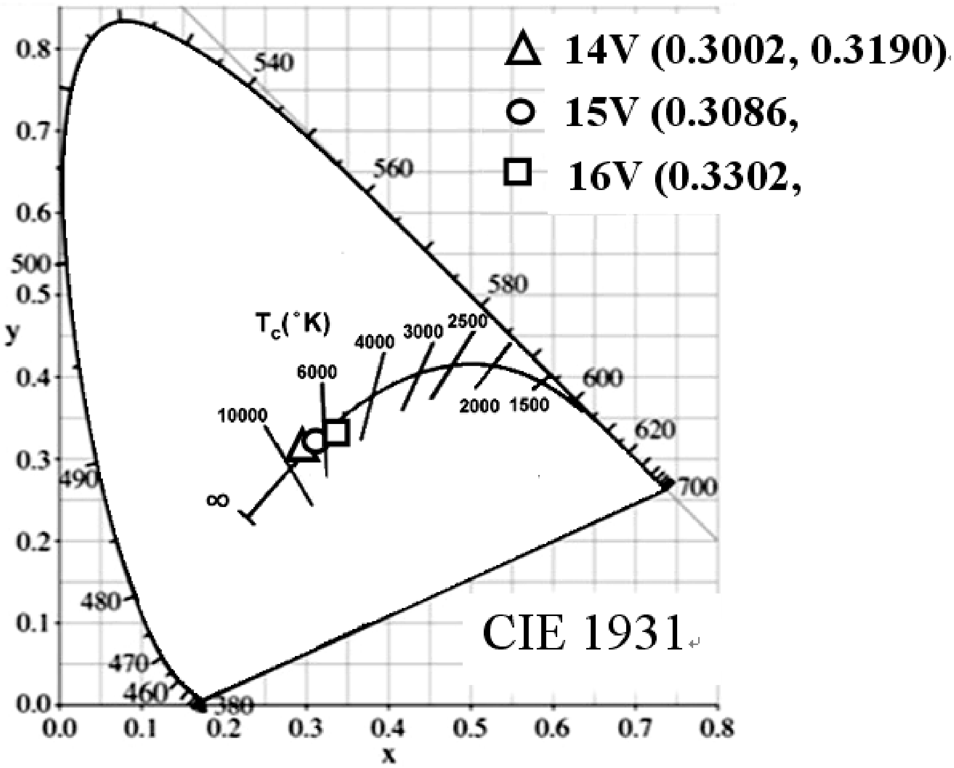
Fig. 2

(a)



Intensity (a.u.)
wavelength (nm)
14 V
15 V
16 V

(b)



14V (0.3002, 0.3190)
15V (0.3086,
16V (0.3302,
CIE 1931
Tc(°K)
x
y

Fig. 3

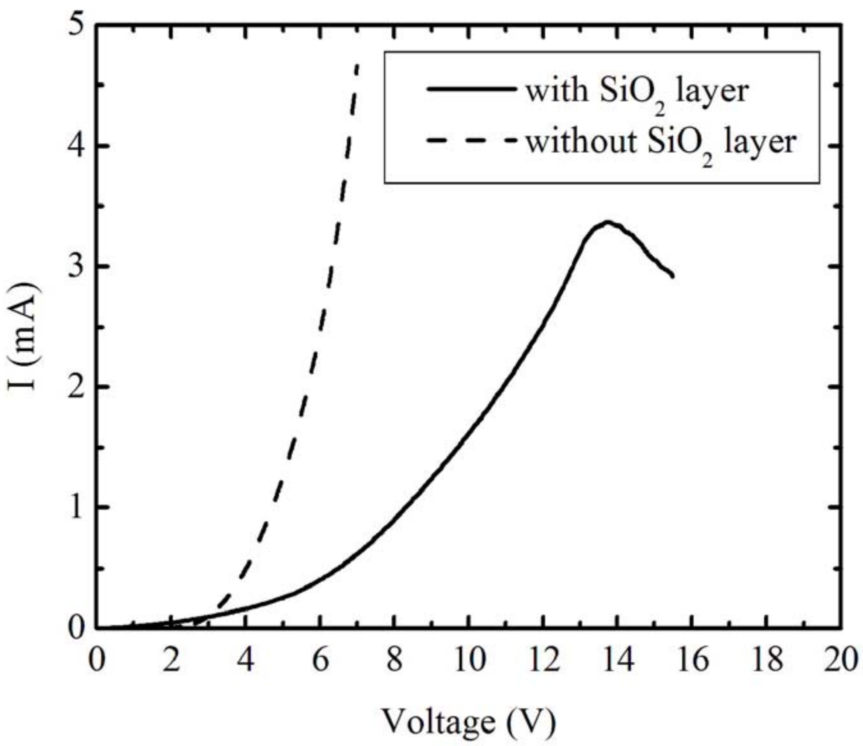


Fig. 4

参考文獻

[1] E. Fred Schubert and Jong Kyu Kim, “Solid-state light sources getting smart,” Science, vol. 308, no. 5726, p 1274-1278, May 27, 2005.
[2] Hao Wu, Xinmin Zhang, Chongfeng Guo, Jian Xu, Mingmei Wu, and Qiang Su, “Three-band white light from InGaN-based blue LED chip precoated with green/red phosphors,“ IEEE Photonics Technology Letters, vol. 17, no. 6, p 1160-1162, June 2005.
[3] J. K. Sheu, S. J. Chang, C. H. Kuo, Y. K. Su, L. W. Wu, Y. C. Lin, W. C. Lai, J. M. Tsai, G. C. Chi, and R. K. Wu, “White-light emission from near UV InGaN-GaN LED chip precoated with blue/green/red phosphors,” IEEE Photonics Technology Letters, vol. 15, no. 1, p 18-20, Jan. 2003.
[4] D. Pacifici, E. C. Moreira, G. Franzò, V. Martorino, F. Priolo, and F. Iacona, “Defect production and annealing in ion-irradiated Si nanocrystals,” Phys. Rev. B, vol. 65, 144109, Apr. 2002.
[5] Qiu-yi Ye, Raphael Tsu, Edward H. Nicollian, “Resonant tunneling via microcrystalline-silicon quantum confinement,” Phys. Rev. B, vol. 44, no. 4, p 1806-1811, 1991.
[6] A. Pèrez-Rodrìguez, O. González-Varona, B. Garrido, P. Pellegrino, J. R. Morante, C. Bonafos, M. Carrada, and A. Claverie, “White luminescence from Si+ and C+ ion-implanted SiO2 films,” J. Appl. Phys., vol. 94, no. 1, p 254-262, July 2003.
[7] X. Zhao, O. Schoenfeld, J. Kusano, Y. Aoyagi, and T. Sugano, “Observation of direct transitions in silicon nanocrystallites,” Jpn. J. Appl. Phys., pt. 2, vol. 33, no. 7A, pp. L899-L901, July 1994.
[8] T. Shimizu-Iwayama, K. Fujita, S. Nakao, K. Saitoh, T. Fujita, and N. Itoh, “Visible photoluminescence in Si+-implanted silica glass,” J. Appl. Phys., vol. 75, no. 12, p 7779-7783, June 1994.
[9] P. Mutti, G. Ghislotti, S. Bertoni, L. Bonoldi, G. F. Cerofolini, L. Meda, E. Grilli, and M. Guzzi, “Room-temperature visible luminescence from silicon nanocrystals in silicon implanted SiO2 layers,” Appl. Phys. Lett., vol. 6, pp. 851-853, Feb. 1995.

[10] Gong-Ru Lin, Chun-Jung Lin, Chi-Kuan Lin, Li-Jen Chou, Yu-Lun Chueh, “Oxygen defect and Si nanocrystal dependent white-light and near-infrared electroluminescence of Si-implanted and plasma-enhanced chemical-vapor deposition-grown Si-rich SiO2,” J. Appl. Phys., vol. 97, p 094306-094306-8, Apr. 2005.
[11] G. Franzò, A. Irrera, E. C. Moreira, M. Miritello, F. Iacona, D. Sanfilippo, G. Di Stefano, P. G. Fallica, F. Priolo, “Electroluminescence of silicon nanocrystals in MOS structures,” Appl. Phys. A: Mater. Sci. Process., vol.74, no. 1, p 1-5, Jan. 2002.
[12] J. Valenta, N. Lalic, J. Linnros, “Electroluminescence microscopy and spectroscopy of silicon nanocrystals in thin SiO2 layers,” Optical Materials, vol. 17, no. 1-2, p 45-50, June 2001.
[13] J. H. Son, T. G. Kim, S. W. Shin, H. B. Kim, W. S. Lee, S. Im, J. H. Song, C. N. Whang, K. H. Chae, “Blue and red luminescence from Si ion-irradiated SiO2/Si/ SiO2 layers,” Optical Materials, vol. 17, no. 1-2, p 125-129, June 2001.

出席「國際電化學年會」及發表論文心得報告

Ecs (electrochemical society) is an international nonprofit, educational organization concerned with a broad range of phenomena relating to electrochemical and solid-state science and technology. The conference's objectives are

1. To advance the theory and practice of electrochemistry, solid-state science, and allied subjects;
2. To encourage research and dissemination of knowledge in these fields; and
3. To assure the availability of adequate training and education of fundamental and applied scientists and engineers in these fields

In this conference, we submitted two oral papers. One is related to ZnO TFTs and the other is about pentacene organic TFTs.

Abstract of this talk is

ZnO-based thin film transistors (TFTs) fabricated on both glass and flexible substrates are investigated. The ZnO thin film mentioned in this entire work is deposited by RF magnetron sputtering at low deposition rate and low temperature.

For glass substrates, we demonstrate a high-performance enhancement-mode ZnO TFT. The I_{DS} is as high as 1 mA when biased at the saturation region V_{DS}=10–20 V and V_{GS}=5 V with a TFT gate size W/L=600μm /300μm. The I_{on}/I_{off} ratio is 3×10^{6}. The results are among the best ZnO TFTs ever obtained. Furthermore, we study the effects of oxygen passivatioin on the ZnO TFT channel layer. Such passivation is able to compensate the oxygen vacancies of the ZnO crystal and to adjust channel carrier concentrations, carrier mobility, crystalline quality, and accordingly TFT performance. Based on the information of the oxygen passivation, we then fabricate top-gate ZnO TFTs with various passivation durations. The performance of ZnO TFTs is closely related to material characterizations.