富含奈米矽晶之過矽二氧化矽發光材料與元件

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摘　　要

本論文旨在於探討過矽二氧化矽材料之製作與材料結構、物性、光性、電性等，並發展含有奈米矽晶之矽基發光二極體。主要可分為五大部分。第一部分為研究 銀電極/矽離子佈植二氧化矽/n 型矽基板/銀電極 金氧半發光二極體之白光、藍綠光，電激螢光與矽離子佈植二氧化矽材料特性。由於大量的矽離子轟擊造成二氧化矽層中許多斷鍵與大量的氧缺陷，在 1100^{o}C 爐管退火 180 分鐘後，主要有三種發光因子，弱氧鍵缺陷(415 nm)，中性氧缺陷(455 nm)與 E’$_{\delta}$ 缺陷(520 nm)。此金氧半發光二極體之電激螢光光譜，會隨不同的逆向偏壓而改變，其是由於反轉層中大量累積的電洞，注入不同能量的發光缺陷因子所造成。在 1.25 安培脈衝式電流驅動下，可達到白光放光功率為 60 nW。第二部分，利用電漿輔助化學氣相沉積法，以不同反應氣體比例(N_2O/SiH_4)製作過矽二氧化矽材料。在 1100^{o}C 退火 30 分鐘後，可得到 4 nm 直徑的奈米矽晶，並製作 銦錫氧透明電極/過矽二氧化矽/p 基板/鋁電極 金氧半發光二極體。利用改變反應電漿功率與基板溫度的方法，探討不同濃度的過矽二氧化矽材料的形成機制。過矽二氧化矽材料易形成於高電漿反應功率與低基板溫度，或低電漿反應功率與高基板溫度。第三部份，由於爐管無法局部區域性退火，故利用二氧化矽材料對二氧化碳雷射波長具有高吸收係數的特性，來進行快速雷射退火，最佳的雷射退火功率為 6 kW/cm^{2}，退火時間為 1 毫秒。過大的雷射功率會造成過矽二氧化矽膜的剝離，與形成中性氧缺陷放光因子，並且增加吸收係數與降低含奈米矽二氧化矽膜之折射率。此

含有奈米矽晶之金氧半發光二極體的載子傳輸機制主要為Fowler-Nordheim穿隧效應。奈米矽晶濃度的增加，使得 銦銻氧透明電極/過矽二氧化矽 之界面位障明顯由3.7 eV降低至1.1 eV，Fowler-Nordheim閾值電場可由2 MV/cm降低至1.4 MV/cm。二氧化碳快速雷射退火之奈米矽晶金氧半發光二極體的發光功率，在電流密度2.3 mA/cm^{2}下，可達50 nW輸出光功率。

第四部份，利用電漿輔助化學氣相沉積法，在低電漿反應功率與高基板溫度下，於過矽二氧化矽膜與矽基板介面，會形成底部約20 nm，高度約10 nm的矽奈米錐。此矽奈米錐可明顯降低Fowler-Nordheim穿隧閾值電場，並增強奈米矽基金氧半發光二極體的發光強度達150 nW。第五部分，利用鎳奈米點為乾式蝕刻檔層，可製作直徑約30 nm，高約350 nm，密度約2.8×10^{10} cm^{-2}之矽奈米柱。此矽奈米柱之不平整性可破壞全內反射，並降低Fowler-Nordheim穿隧電場，增加注入矽奈米晶體的載子，使得在375微安培電流驅動下，可達0.7微瓦的光功率輸出，並可達到0.1 %的外部量子效率。

Silicon-rich silicon dioxide light emitting material and device with buried nanocrystallite silicon

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ABSTRACT

This dissertation explores the fabrication, structural properties, physical features, optical and electronic properties of Si-rich SiO_2 (SiO_x) material and light emitting device with buried nanocrystallite silicon. The main focus of this dissertation can be divided into five parts.

First, the white-light and blue-green electroluminescence (EL) from an Ag/ Si-ion-implanted SiO_2 (SiO_2:Si^+)/n-Si/Ag metal-oxide-semiconductor light emitting diode (MOSLED) with the defect-enhanced blue-green photoluminescence (PL) are studied. After annealing for 180 min at 1100^{o}C, the main irradiative defects corresponding to PL at 415, 455 and 520 nm are completely activated, which are identified as weak oxygen bond (WOB), the neutral oxygen vacancy (NOV)-related defects and E'_{δ}-related defects, respectively. During the Si implantation (or physical bombardment with high-energy ions), the oxygen vacancies and the oxygen interstitials (the precursors for the WOB defects) are created due to the relatively large quantities of oxygen that are displaced from their atomic positions in the SiO_2 matrix. The EL spectrum of the MOS diode under different reverse bias conditions indicates that the irradiative recombination is due to enhanced impact ionization of ground states of defects, such as WOB, NOV, and E'_{δ} defects, through the injection of holes accumulated in the inversion layer formed beneath the SiO_2:Si^+/n-Si interface. The maximum white-light luminescent power is up to 60 nW at a pulsed bias current of 1.25 A.

Next, the 4-nm Si nanocrystal (nc-Si) contributed to PL and EL at about 760 nm is precipitated in the plasma enhanced chemical vapor deposition (PECVD)-grown Si-rich SiO_x film after annealing at 1100 °C for 30 min. Under a high RF power condition, the increasing substrate temperature usually inhibits the precipitation of nc-Si since high-temperature growth facilitates stochiometric SiO_2 deposition. The indium-tin-oxide (ITO)/PECVD-grown SiO_x/p-Si/Al MOS diode is highly resistive with turn-on voltage and power-current (P-I) slope of 86 V and 0.7 mW/A, respectively. The decomposed EL peaks at 625 and 768 nm are contributed by the bias-dependent cold-carrier tunneling between the excited states in adjacent nc-Si quantum dots. Both evaluated ITO/SiO_x junction barrier height and Fowler-Nordheim (FN) tunneling threshold decreased from 3.7 to 1.1 eV and from 2 to 1.4 MV/cm, respectively. Low-plasma-power and oxygen-deficient deposited

SiO_x at 400°C provides nc-Si density of 6.4×10^{18} cm^{-3}, enhancing internal quantum efficiency and power-current slope of an ITO/SiO_x/p-Si/Al MOSLED by one order of magnitude.

Third, Structural damage enhanced near-infrared EL of a MOSLED made on SiO_x film with buried nanocrystallite Si after CO_2 laser rapid-thermal-annealing (RTA) at an optimized intensity of 6 kW/cm^2 for 1 ms is demonstrated. Laser ablation is initiated at a laser intensity of >7.5 kW/cm^2, leaving numerous luminescent centers that are related to NOV defects, increasing the absorption coefficient and related optical bandgap energy, and reducing the refractive index in partially annealed SiO_x. CO_2 laser RTA induced oxygen-related defects are capable of improving FN tunneling mechanism of carriers at metal/SiO_x interface. The CO_2 laser RTA SiO_x film reduces FN tunneling threshold to 1.8 MV/cm, facilitating an enhanced EL power of an ITO/SiO_x/p-Si/Al MOSLED up to 50 nW at a current density of 2.3 mA/cm^2.

Fourth, the premier observation on the enhanced light emission from such an ITO/SiO_x/p-Si/Al MOSLED with Si nano-pyramids at SiO_x/Si interface is demonstrated at low biases. The Si nano-pyramids exhibits capability in providing the roughness of the SiO_x/Si interface, and improving the F-N tunneling mechanism based carrier injection through the novel SiO_x/nano-Si-pyramid/Si structure. With these Si nano-pyramids at a surface density of up to 10^{12}/cm^2, the F-N tunneling threshold can be reduce from 7 MV/cm to 1.4 MV/cm. The correlation between surface density of the interfacial Si nano-pyramids and the threshold F-N tunneling field has been elucidated. An output EL power of nearly 150 nW under a biased voltage of 75 V and current density of 32 mA/cm^2 is reported for the first time.

The last one is that nc-Si based MOSLED on Si nano-pillar array with size, height and density of 30 nm, 350 nm and 2.8×10^{10} cm^{-2}, respectively, is characterized. The nano-roughened Si surface contributes to both the relaxation of total-internal reflection at device-air interface and the FN tunneling enhanced turn-on characteristics, providing the MOSLED a maximum optical power of 0.7 μW obtained at biased current of 375 μA. The optical intensity, turn-on current and power-current slope of nc-Si MOSLED on high-aspect-ratio Si nano-pillar array are 140 μW/cm^2, 5 μA, 2±0.8 mW/A, respectively. A maximum external quantum efficiency of up 0.1% is reported.