Enhanced performance of silicon quantum dot light-emitting diodes grown on nanoroughened silicon substrate

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We report the effect of a nanoroughened Si substrate on silicon quantum dot (Si QD) light-emitting diodes (LEDs). The electroluminescence of Si QD LEDs grown on the nanoroughened Si substrate was remarkably improved by 493% at an injection current of 90 mA compared to those of Si QD LEDs grown on the flat Si substrate. The electrical and optical enhancements were attributed to the enhanced inhomogeneous local electric field on the nanoroughened Si surface and the angular randomization of photons emitted from Si QDs at the nanoroughened surface of silicon nitride layer containing Si QDs. © 2009 American Institute of Physics. [DOI: 10.1063/1.3211113]

Silicon quantum dots (Si QDs) have attracted considerable interest due to their potential as light sources for integrated silicon photonics.1 Although the electroluminescence (EL) intensity of Si QD light-emitting diodes (LEDs) continues to increase, the external quantum efficiency (which is a product of the radiative efficiency, current injection efficiency, and extraction efficiency)2) is still too low for many practical applications.3 11 One of the main reasons for low external quantum efficiency is the use of an insulator matrix with a large tunneling barrier height. It is very difficult for charge carriers to flow from metal contact layers to the QDs when the QDs are embedded in silicon nitride or silicon oxide films. Furthermore, the light-extraction efficiency of the LEDs is limited by total internal reflection of the light at the interface between the active layer and air. To obtain high efficiency QD LEDs, it is very important to increase the tunneling current from the metal contact and the light-extraction efficiency of the overall structure.12 11 In this letter, we report the enhancement of electrical and optical properties of Si QD LEDs obtained by growing them on Si substrates which were nanoroughened using a wet etching process.

Nanoroughened Si surfaces were formed on p-type Si wafers (100) with a hole concentration of approximately 1015 cm−3. The wet etching was accomplished using 4.5 and 13.5 wt % KOH solutions at room temperature. The 40-nm-thick silicon nitride films containing Si QDs were grown on the Si wafer using plasma-enhanced chemical vapor deposition with NH3 and nitrogen-diluted 5% SiH4 reactant gases.17 19 Unalloyed metal contacts consisting of Ni (9 nm) and Au (21 nm) were deposited on the silicon nitride films using e-beam evaporation. A transparent NiO film was formed by annealing the Ni/Au contact at 400 °C for 80 s in air. A Pt (20 nm)/Au (100 nm) contact was then deposited on the back side of the Si substrate. A Si QD LED with a flat Si surface was fabricated for comparison.

Figure 1 contains atomic force microscope (AFM) images of the Si surface. Figure 1(a) is an AFM image of the flat Si surface and Figs. 1(b) and 1(c) are AFM images of the surfaces of Si wafers etched by immersion in 4.5 or 13.5 wt % KOH solutions for 10 min. The root mean square (rms) roughness of the flat Si surface is 0.7 Å, while the Si surfaces etched by 4.5 or 13.5 wt % KOH solutions have roughness values of 73 or 191 Å. The average aspect ratio (height-to-radius) of the nanoprotrusions on the flat Si surface was estimated to be 0.005, while nanoprotrusions on Si

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FIG. 1. AFM images presenting the surface morphologies of (a) flat Si surface and Si surfaces nanoroughened by using (b) 4.5 wt % and (c) 13.5 wt % KOH solutions, respectively, where rms roughnesses of Si surfaces were 0.7, 73, and 191 Å.
The local electric field is related to the concentration of the KOH etching solutions. In a previous study, the use of different substrates with different degrees of roughness did not affect the concentration of QDs that formed within amorphous silicon nitride films during deposition on these surfaces via plasma-enhanced chemical vapor deposition. Therefore, we believe that the differences in the concentrations of QDs deposited on nanoroughened and flat Si surfaces will be essentially the same.

Figure 2(a) depicts the current-voltage (I-V) curves of the Si QD LED with a flat Si surface and the Si QD LEDs with Si surfaces nanoroughened by using 4.5 and 13.5 wt % KOH solutions, which display threshold voltages of 13.4, 12.7, and 9.8 V, respectively. The threshold voltage of Si QD LEDs with nanoroughened Si surfaces was decreased by 3.6 V compared to the reference LED. To understand the low threshold voltage of nanoroughened Si QD LEDs, the Fowler–Nordheim (FN) current densities of a Si QD LED with a flat Si surface and Si QD LEDs with roughened Si surfaces were plotted in Fig. 2(b). The FN tunneling current density \( J_{\text{FN}} \) is given by \( J_{\text{FN}}=AE^2 \exp(-B/E) \), where \( A=\sqrt{q/m^*} / (8\pi\hbar^2\Phi_p) \), \( B=(8\pi^2/m^*q^2\Phi_p) / (3q\hbar) \), in which \( E \) is the local electric field, \( m \) and \( m^* \) are the free electron mass and the effective mass in the silicon nitride, \( \Phi_p \) is the potential barrier height between silicon and silicon nitride, and \( h \) is Planck’s constant. The local electric field is related to the applied voltage \( V \) by \( E=\beta V/d \), where \( d \) is the thickness of the silicon nitride film and \( \beta \) is an enhancement factor of the local electric field dependent on the aspect ratio and density of protrusions on the Si substrate. The enhancement factor \( \beta \) can be obtained by plotting \( \ln(J/E^2) \) versus \( 1/E \) and determining the inverse of the slope. The flat surfaced LED exhibited a steeper FN slope [Fig. 2(b)], and \( \beta \) increased with increasing rms roughness of the Si wafer. The increased tunneling probability of carriers through the roughened Si surface is attributed to enhancement of the local electric field. Therefore, the injection efficiency of carriers from the substrate into the QDs can be greatly increased by using roughened Si surfaces as QD LED substrates.

Figure 3 compares the EL and photoluminescence (PL) spectra of the reference LED and the nanoroughened Si QD LEDs. The EL peak positions of the roughened LEDs were similar to the PL peak position of the reference LED, indicating that the origins of the EL and PL are identical. However, the high-energy side of the EL peak decreased in devices etched at a higher KOH concentration. This is attributed to the decrease in the recombination probability of decreased carriers in small Si QDs as the injection current density decreases. The EL intensities of the nanoroughened Si QD LEDs were remarkably enhanced compared to the reference LED, mostly due to the enhanced current injection efficiency and the increased probability of carriers tunneling from the substrate into the QDs, as described in Fig. 2. The large EL enhancement can also be attributed to increased light-extraction efficiency. In the reference LED, the light-extraction efficiency is estimated to be 6.1% from \( P_{\text{air}}/P_{\text{source}}=(1/4)(n_{\text{air}}^2/n_{\text{Si3N4}}^2) \), where \( P_{\text{air}} \) is the emitted power, \( P_{\text{source}} \) is the power generated by the LED, and \( n_{\text{Si3N4}} \) and \( n_{\text{air}} \) are the refractive indices of Si3N4 (2.02) and air (1.00) at a wavelength of 620 nm. The low light-extraction efficiency is due to the small critical angle of 29.6° estimated from Snell’s law, \( \theta_a=\sin^{-1}(n_{\text{air}}/n_{\text{Si3N4}}) \). In Si QD LEDs with a nanoroughened Si surface, the escape probability of photons generated inside the QDs is expected to be increased due to angular randomization. Figure 4 depicts the integrated EL intensities of the Si QD LEDs in the visible range as a function of the injection current. The EL intensities of Si QD LEDs roughened using 4.5 wt % KOH were enhanced by 203% and the intensities of those roughened with 13.5 wt % KOH were enhanced by 493% at a forward current of 90 mA. The large EL intensities displayed by Si QD LEDs with nanoroughened Si surfaces are attributed both to increased current injection efficiency and to increased light-extraction efficiency.
In summary, we report an enhancement of the electrical and optical properties of Si QD LEDs by fabricating them on a nanoroughened Si surface. The EL emission was increased by 493% at a current of 90 mA compared to a reference LED. The improved electrical properties were attributed to a locally enhanced electric field at the irregularities on the Si surface. The nanoroughened surface structures further improved the escape probability of photons due to angular randomization of the emission, resulting in an increase in the light-extraction efficiency.

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