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Electroluminescence from Au/(SiO_2 /Si/SiO_2) nanometer double barrier/p-Si structures and its mechanism

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Abstract

SiO_2/Si/SiO_2 nanometer double barriers (SSSNDB) with Si layers of twenty-seven different thicknesses in a range of 1–5 nm with an interval of 0.2 nm have been deposited on p-Si substrates using two-target alternative magnetron sputtering. Electroluminescence (EL) from the semitransparent Au film/SSSNDB/p-Si diodes and from a control diode without any Si layer have been observed under forward bias. Each EL spectrum of all these diodes can be fitted by two Gaussian bands with peak energies of 1.82 and 2.25 eV, and full widths at half maximum of 0.38 and 0.69 eV, respectively. It is found that the current, EL peak wavelength and intensities of the two Gaussian bands of the Au/SSSNDB/p-Si structure oscillate synchronously with increasing Si layer thickness with a period corresponding to half a de Broglie wavelength of the carriers. The experimental results strongly indicate that the EL originates mainly from two types of luminescence centres with energies of 1.82 and 2.25 eV in the SiO_2 barriers, rather than from the nanometer Si well in the SSSNDB. The EL mechanism is discussed in detail.

1. Introduction

Dimaria *et al* [1] first reported electroluminescence (EL) from an Au/SiO_2 (50 nm)/Si-rich Si oxide (20 nm)/n-Si structure annealed at 1000 °C in ambient N_2 under a forward bias

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greater than 15 V. They explained the visible EL using the quantum confinement model: band to band radiative recombination of electron-hole pairs occurs within nanoscale silicon particles with energy gaps larger than that of bulk Si due to the quantum confinement effect in the Si-rich Si oxide. In our previous studies, visible EL from the Au/native Si oxide/p-Si structure [2] and Au/ultrathin Si-rich SiO_2 layer/p-Si structure under a forward bias was reported [3]. Some essential differences between Dimaria's experiment and ours are as follows. In their experiment, the SiO_2 layers were as thick as 70 nm, the substrate was n-Si wafer, and quite strong EL from the Au/SiO_2 (50 nm)/Si-rich Si oxide (20 nm)/n-Si structure was observed only after the SiO_2 (50 nm)/Si-rich Si oxide (20 nm)/n-Si structure was annealed at 1000 °C. However, in our experiments [2, 3], the Si oxide layers were only 3–5 nm thick, much thinner than 70 nm, the substrate was p-Si wafer rather than n-Si wafer (when we used an n-Si substrate, no EL could be observed except under a very high reverse bias), and quite strong EL from the above two structures was observed even without any annealing. Based on our experimental results, we proposed that EL should originate from the radiative recombination of electron-hole pairs via the luminescence centres (defects and impurities) rather than from nanoscale Si clusters in the Si oxide layer. We believe that the main cause of disagreement on the EL mechanism is the fact that the size of the Si clusters in such Si-rich SiO_2 films was nonuniform and so very difficult to measure exactly. Thus, it was difficult to determine whether luminescence originated from the nanoscale Si clusters or from luminescence centres in the Si oxide layer. To overcome this obstacle, we recently studied the EL from an Au/(SiO_2 /Si/SiO_2) nanometer double barrier (SSSNDB)/p-Si sandwich structure with a nanometer Si layer of uniform and controllable thickness, instead of nanoscale Si clusters in a Si-rich SiO_2 layer as in the previously studied Au/ultrathin Si-rich SiO_2 layer/p-Si structure [4]. In this paper we report the variation of current of the sandwich structure with increasing Si layer thickness, and show that each EL spectrum of all the Au/SSSNDB/p-Si diodes and the control diode without any Si layer in the SSSNDB can be decomposed into two Gaussian bands with certain peak energy and full-width-at-half-maximum (FWHM) values. The intensities of the two Gaussian bands vary synchronously with the current when the Si layer thickness increases. This variation is considered to be the cause of the variation of the intensities of the two Gaussian bands. It is seen that the current oscillation period corresponds to half a de Broglie wavelength of the carriers, which can be explained by the transfer matrix theory. The EL mechanism of the structures studied is discussed in detail.

2. Experiment

The SSSNDB structure was deposited on a (100)-oriented, 5–9 cm, p-type Si substrate. The thickness of the nanometer Si layer in the SSSNDB varied from 1 to 5 nm with an interval of 0.2 nm, while the thicknesses of the upper (near the Au electrode) and lower (near the p-Si substrate) SiO_2 layers in all the samples were 3.0 and 1.5 nm, respectively. A control structure without any Si layer, i.e., a single SiO_2 layer with a thickness of 4.5 nm instead of SSSNDB, was also deposited on a p-Si substrate. The deposition technology was the same as depicted in [4]. After deposition, all the SSSNDB/p-Si and control samples were annealed at 600 °C for 30 minutes in N_2 . Finally, semitransparent Au films 3 mm in diameter were deposited on the samples' front surfaces as electrodes.

3. Results

All the samples, including the control diode, exhibit good rectifying junction behaviour. For clarity, only the $I - V$ characteristic curves of the selected five sample diodes and the control diode are shown in figure 1. Visible light from both the investigated and control structures can be observed by the naked eye when the forward bias exceeds 6 V and becomes even stronger with increasing bias. Under a reverse bias, however, no EL has been observed. The EL spectra of the control diode and thirteen other diodes with Si layers of different thicknesses are shown in figure 2. Figures 3(a), (b) and (c), show, respectively, the current, EL intensity and EL peak wavelengths as functions of the Si layer thickness. It can be seen that:

(a) The current, EL intensity and EL peak wavelength of the investigated structure oscillate synchronously with increasing Si layer thickness. The oscillation period is about 0.4–0.6 nm.
(b) EL is observed in the control structure as well, although there is no Si layer in the structure. The current, EL intensity and EL peak wavelength of the control diode all have a value between the maximum and minimum (close to the latter) of the corresponding quantities of the investigated diodes.
(c) If the SiO_2 (4.5 nm) layer in the control structure is removed, no EL can be observed from the resultant Au/p-Si structure.

Because all the EL spectra of the diodes have a peak between 1.8 and 2.0 eV and a shoulder of $\sim$ 2.2 eV, we try to fit them with two Gaussian bands with fixed peak energies and FWHMs. The results show that they can be fitted well by two Gaussian curves with peak energies of 1.82 and 2.25 eV and FWHMs of 0.38 and 0.69 eV, respectively. Figure 4 shows, for example, how the EL spectrum of six investigated diodes with Si layers having different thicknesses and that of the control diode are decomposed into two such Gaussian bands. Figure 5 shows the current and intensities of the two Gaussian bands for the investigated diodes, as well as for the control diode for comparison, as functions of the Si layer thickness. It is found that the intensities of the two Gaussian curves of the investigated structure vary synchronously with the current as the Si layer thickness increases.

Although most of the EL spectra of the investigated diodes are evidently different from that of the control diode, the EL spectra of the investigated diodes with Si layers of thickness 1.4, 2.0 and 2.2 nm are similar to that of the control diode, as shown in figure 6.

4. Discussion

If the quantum confinement model were appropriate for explaining the origin of the EL, the following predictions could be made. (a) The EL peak wavelength of the investigated diodes should monotonically redshift with increasing Si layer thickness. (b) The EL intensity of the investigated diode should monotonically decrease with increasing Si layer thickness. (c) No or very weak EL should be observed from the control diode because there is no or little nanometer Si layer present. (d) Even if the control diode had a few nanoscale Si clusters, its EL spectrum should be markedly different from that of the investigated diodes because they have different EL origins: a few nanoscale Si clusters in the Si oxide layer of the control diode as compared to a nanoscale Si layer sandwiched between two Si oxide layers in the investigated diode. However, all these predictions are contrary to what we have observed. In fact, (a) the EL peak wavelength of the investigated diode oscillates rather than monotonically redshifts with increasing Si layer thickness; (b) the EL intensity of the investigated diode oscillates rather than monotonically decreases with increasing Si layer thickness; (c) quite strong EL

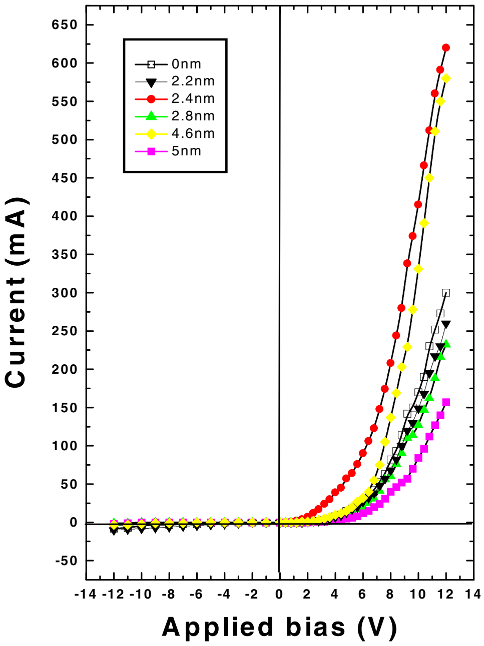

Figure 1. $I - V$ characteristic curves of five selected Au/SSSNDB/p-Si diodes with Si layers of thickness 2.2, 2.4, 2.8, 4.6 and 5.0 nm, and of an Au/SiO$_2$(4.5 nm)/p-Si diode.

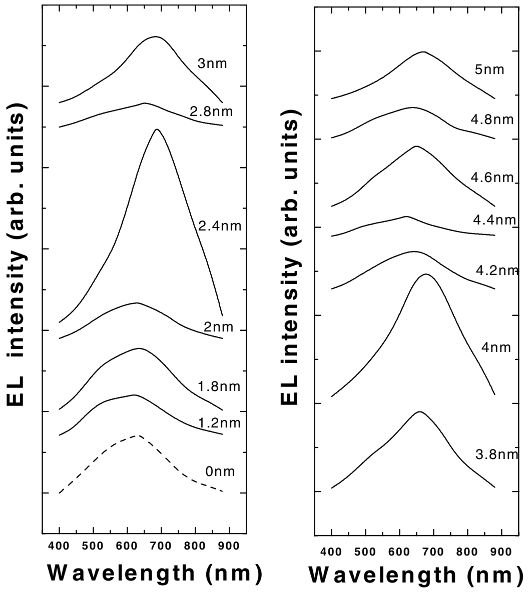

Figure 2. Thirteen EL spectra of the Au/SSSNDB/p-Si diodes with Si layers of thickness 1.2, 1.8, 2.0, 2.4, 2.8, 3.0, 3.8, 4, 4.2, 4.4, 4.6, 4.8 and 5.0 nm (solid lines), and EL spectrum of the Au/SiO$_2$ (4.5 nm)/p-Si diode (dashed line), all under a forward bias of 8 V.

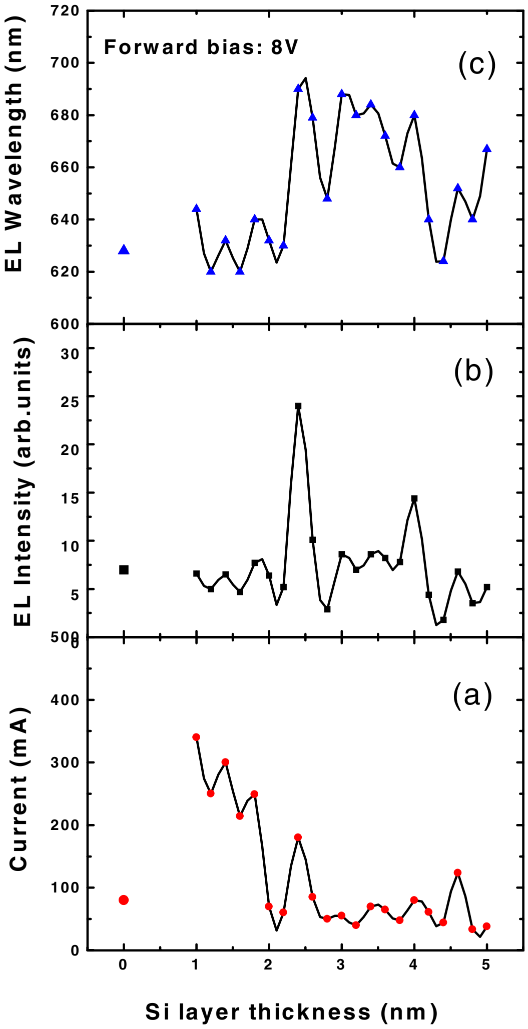

Figure 3. (a), (b) and (c), show, respectively, the currents, EL intensities and peak wavelengths of the Au/SSSNDB/p-Si diodes under a forward bias of 8V as functions of Si layer thickness in the range 1–5 nm. The same parameters of the control diode corresponding to a zero Si layer thickness under the same bias are shown as solid circles, solid squares and solid triangles, respectively.

can be observed from the control diode, and its intensity is even stronger than that of some investigated diodes, as shown in figure 3(b); (d) The EL spectra of some investigated diodes are similar to the EL spectrum of the control diode, though a nanoscale Si layer exists only in the investigated diodes and not in the control one.

To interpret these experimental results we suggest the following EL model for both the investigated and control structures. (1) EL originates from radiative recombination of electron-hole pairs via two types of luminescence centres (LCs) with luminescence energies of 1.82 and 2.25 eV, which we shall refer to as LC1 and LC2, respectively, in the SiO_2 layers. (2) There are four types of transport-recombination processes in the investigated structure shown in figure 7: (A) Electrons from the Au electrode and holes from the p-Si substrate tunnel into the nanometer Si layer, relax into the lowest unoccupied molecular orbit and the highest occupied

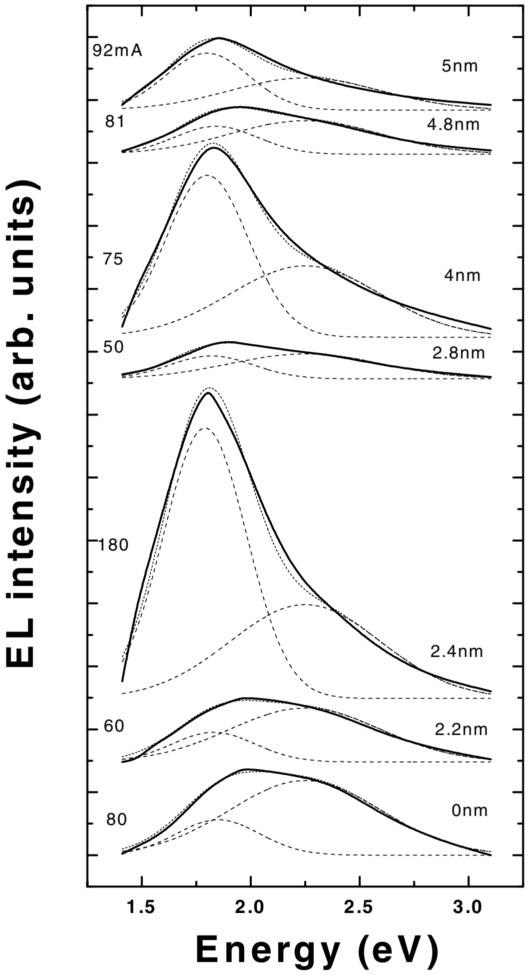

Figure 4. EL spectra of six Au/SSSNDB/p-Si diodes with Si layers of thickness 2.2, 2.4, 2.8, 4.0, 4.8 and 5.0 nm and of the Au/SiO_2 (4.5 nm)/p-Si diode (all under a forward bias of 8 V), decomposed into two Gaussian bands with peak energies of 1.82 and 2.25 eV and FWHMs of 0.38 and 0.69 eV, respectively. The two Gaussian bands used in each fitting are depicted by dashed lines, and the fitted curves by dotted lines.

molecular orbit, respectively, then directly or indirectly (through deep levels) recombine in the Si layer. Process A is just the process described in the quantum confinement model for EL; (B) Electrons and holes tunnel directly into LCs in the SiO_2 layers, then radiatively or nonradiatively recombine there. This process B is referred to as the tunneling-luminescence centre process; (C) Electrons and holes tunnel into the nanomater Si layer first. During or after relaxing into the lowest unoccupied molecular orbit and highest occupied molecular orbit, electrons and holes, respectively, tunnel into the LCs in SiO_2 layers, then radiatively or nonradiatively recombine there; (D) electrons (holes) tunnel from the Au electrode (p-Si substrate) into the LCs in the SiO_2 layers, whereas holes (electrons) tunnel from the p-Si substrate (Au electrode) into the Si layer first and then into the LCs in the SiO_2 layers and recombine there with the electrons (holes). Processes C and D are referred to as tunneling-

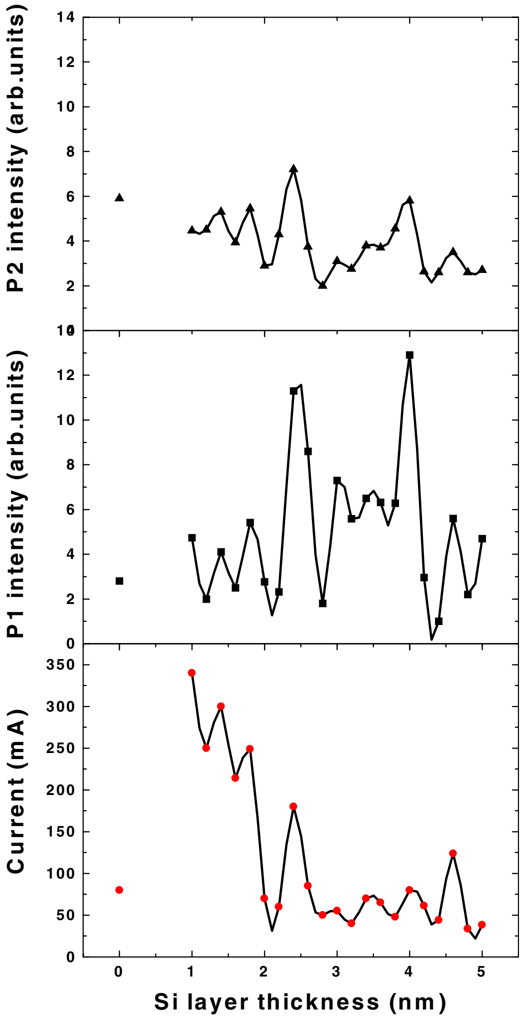

Figure 5. Current and intensity of the two Gaussian bands of the Au/SSSNDB/p-Si diodes under a forward bias of 8 V, as functions of increasing Si layer thickness in the range 1–5 nm.

quantum confinement-luminescence centre processes. We consider that process A really exists, but that processes B, C and D are the major ones in the investigated structure, while process B is the only major transport-recombination process in the control structure.

Based on the suggested EL model and the fact that current varies with Si layer thickness, which can be interpreted by the transfer matrix theory and will be discussed later, all the experiment results can be qualitatively explained as follows.

(a) The fact that each EL spectrum of the investigated diodes irrespective of the thickness of the nanometer Si layer can be decomposed into two Gaussian bands with definite peak energies and definite FWHMs is a direct result of the supposition that EL originates mainly from two types of LCs in the SiO_2 layers.

(b) As the Si layer thickness increases, the oscillation of EL intensity and peak wavelength can be explained by the variation of the current as follows. When resonant tunneling occurs,

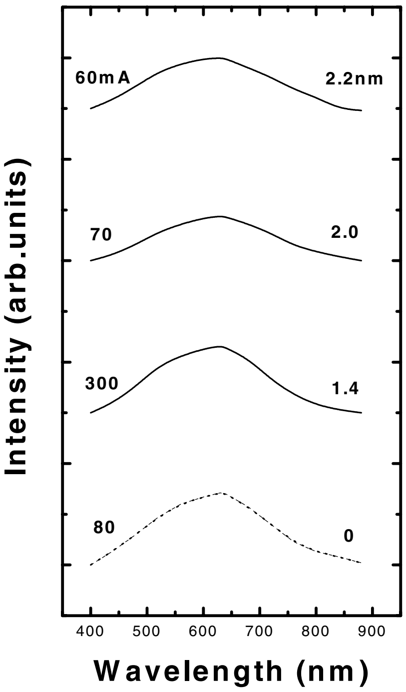

Figure 6. Some EL spectra of the Au/SSSNDB/p-Si diodes with Si layers of thickness 1.4, 2.0 and 2.2 nm (solid lines), and of the control diode (dashed line) under a forward bias of 8 V.

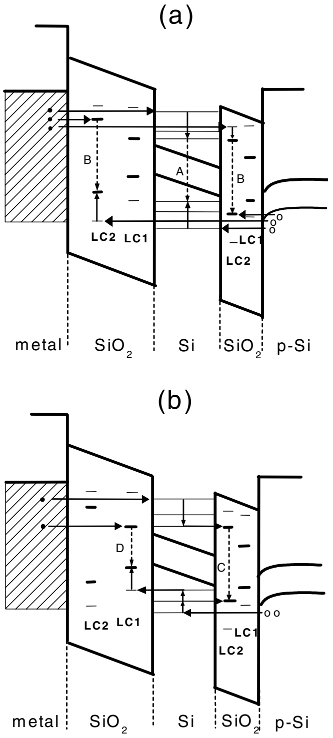

Figure 7. (a) and (b) Schematic of the four types of transport-recombination processes in an Au/SSSNDB/p-Si structure with two types of LCs in the SiO_2 layers under a forward bias. Processes (A)-(D) are explained in the text.

the current reaches a maximum and the respective densities of the electrons and holes in the nanometer Si well also reach a maximum. Then the probability of electrons and holes tunneling into the two types of LCs in the SiO_2 layers via processes C and D, and thus the intensities of the two Gaussian bands corresponding to radiative recombination in LC1 and LC2 also, attain their maxima. As a result, the intensity of the EL peak, which is a sum of the two Gaussian bands, attains a maximum. Because the intensities of the two Gaussian bands synchronously swing with increasing Si layer thickness, so does the EL intensity, as shown in figure 3. As the Si layer thickness increases, the current increases or decreases, so if the enhanced or reduced proportions of the luminescence intensities of the two LCs were the same, the EL peak wavelength would remain invariable. In fact, when the Si layer thickness increases, for example from 2.2 to 2.4 nm, the current increases from $\sim$ 60 to $\sim$ 180 mA and the peak shifts from $\sim$ 630 to $\sim$ 690 nm. The fact that it shifts to a longer value can be explained by the fact that the enhanced proportion of the luminescence intensity from the LC with a lower energy is larger than that from the LC with a higher energy. Thus, when the Si layer thickness increases continuously, the current variation will result in synchronized variation of the peak wavelength as well as the intensity.

(c) The fact that quite strong EL from the control structure is observed and its spectrum can also be decomposed into the two Gaussian bands mentioned above is because EL originates mainly from the two types of LCs in the SiO_2 layers and because the transport-recombination process B is one of the major processes. The reason why the EL intensity of the control diode is larger than that of some of the other diodes may be ascribed to the fact that the energy levels of carriers in the nanometer Si wells of the latter are not aligned with those of the LCs in the nanometer SiO_2 barriers.

(d) Although the transport-recombination processes in the investigated and control structures are different, the main EL sources are the same, thus their spectra are similar.

(e) The experimental result that EL still can be observed when the Si layer is absent from the semitransparent Au/SSSNDB/p-Si structure, but not when the SiO_2 (4.5 nm) layer is removed before semitransparent Au film deposition, is direct evidence that EL originates from the LCs in the SiO_2 barriers rather than from the Si well.

As for the origin of the two types of LCs, we consider that for the 1.82 eV band, the corresponding LC may arise from the nonbridging oxygen hole centre, which is an important LC in Si oxide and is reported to have an emission energy around 1.9 eV [5]. The LC responsible for the 2.25 eV band is probably an oxygen-surplus-type defect [6] or an E'_δ centre [7], which have light emission energies of about 2.25 or 2.2 eV.

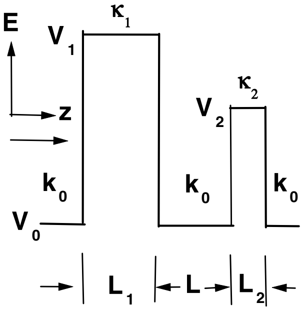

Figure 8. A general one-dimensional asymmetric double-barrier single quantum well structure along the z axis.

In the carrier transport process, due to the interference effect of electron waves within the SSSNDB double-barrier structure, the current as a function of Si layer thickness attains a maximum at a series of Si layer thicknesses which is known as resonant tunneling [8, 9]. To understand this further we can consider the carrier transportation in an Au/ asymmetric double-barrier single quantum well /p-Si structure in terms of transfer matrix theory [10–12]. Figure 8 shows schematically a general one-dimensional asymmetric double-barrier single quantum well structure along the z axis, where V_0, V_1 and V_2 are potential energies for carriers in different regions. We suppose $V_0 < E_z < V_1$ or V_2, where E_z is the carrier energy in the z direction and the transmission coefficient T can be obtained as:

$$T = \frac{1}{1 + \frac{1}{T_1 T_2}[R_1 + R_2 - 2R_1R_2\cos(2k_0L + \alpha_1 + \alpha_2)]}. \tag{1}$$

Here L is the width of the Si well, and T_i and R_i ($i = 1$ and 2) are the transmission and reflection coefficients for a single barrier, respectively, and are given by:

$$T_i = \frac{1}{1 + \frac{1}{4}(\frac{\kappa_i}{k_0} + \frac{k_0}{\kappa_i})^2 \mathrm{sh}^2(\kappa_i L_i)} \tag{2}$$

$$R_i = \frac{\frac{1}{4}(\frac{\kappa_i}{k_0} + \frac{k_0}{\kappa_i})^2 \mathrm{sh}^2(\kappa_i L_i)}{1 + \frac{1}{4}(\frac{\kappa_i}{k_0} + \frac{k_0}{\kappa_i})^2 \mathrm{sh}^2(\kappa_i L_i)} \tag{3}$$

where

$$k_0 = \frac{\sqrt{2m^*(E_z - V_0)}}{\hbar} \tag{4}$$

$$\kappa_i = \frac{\sqrt{2m^*(V_i - E_z)}}{\hbar} \tag{5}$$

$i = 1$ or 2, m^* is the carrier effective mass, h is Planck's constant, and α_i $(i = 1, 2)$ in equation (1) has the following formula:

$$\alpha_i = \mathrm{arcctg}[\frac{1}{2}(\frac{\kappa_i}{k_0} - \frac{k_0}{\kappa_i})\mathrm{th}(\kappa_i L_i)]. \tag{6}$$

Note that in equation (1) only the $\cos(2k_0 L + \alpha_1 + \alpha_2)$ term depends on the Si well width L. The wavevector k_0 equals $2\pi/\lambda$, where λ is the de Broglie wavelength of the carriers. From equation (1) it is found that the current period as a function of L is $\lambda/2$. From equation (4),

$$\lambda = \frac{h}{\sqrt{2m^*(E_z - V_0)}}. \tag{7}$$

Taking $m^* = m_0$ (free electron mass) and $E_z - V_0 = 23$ eV (for a forward bias of 8 eV), $\lambda = 0.7$–1.0 nm is obtained. The current period shown in figure 3(a) varies through a range of 0.4–0.6 nm, which is consistent with $\lambda/2$, half a de Broglie wavelength.

We consider that the EL model suggested for the Au/SSSNSB/p-Si structure is also suitable in principle for the Au/Si-rich Si oxide/p-Si and Au/oxidized porous Si/p-Si structures because they are similar in structure, i.e. they all have Si oxide embedded with nanoscale Si as the active region, and thus have similar EL spectra [2, 3]. (There is a difference, though, in that the nanoscale Si clusters have random sizes in the Si-rich Si oxide and oxidized porous Si, while the nanometer Si layer has uniform thickness in the SSSNDB).

5. Summary

Electroluminescence from Au/SSSNDB/p-Si diodes with Si layers of twenty-seven different thicknesses in a range of 1–5 nm and from a control diode of the Au/SiO$_2$ (4.5 nm)/p-Si structure has been observed and studied. Each EL spectrum of the diodes can be decomposed into two Gaussian bands with peak energies of 1.82 and 2.25 eV and FWHMs of 0.38 and 0.69 eV, respectively. The current and intensities of the two Gaussian bands of the Au/SSSNDB/p-Si structure under a certain forward bias oscillate synchronously with increasing Si layer thickness. The current variation due to the interference of electron waves within the SSSNDB structure is considered to be the cause of the intensity variation of the two Gaussian bands, and as a result, the intensity and peak wavelength of the EL spectra of the structure vary synchronously with increasing Si layer thickness. The period corresponds to half a de Broglie wavelength of the carriers. The experimental results strongly indicate that the EL originates mainly from luminescence centres (LCs) in the SiO$_2$ barriers rather than from the Si well. Two types of LCs in the SiO$_2$ layers with luminescence peak energies at about 1.82 and 2.25 eV play major roles in the EL. In addition, the tunneling process across the Si oxide layer and the quantum confinement effect in the Si well play important roles in the transportation of carriers and thus in the EL.

Acknowledgments

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