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Electroluminescence from silicon nanowires

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Abstract
Room temperature electroluminescence has been demonstrated from undoped silicon nanowires that were grown from disilane. Ensembles of nanowires were excited by capacitively coupling them to an ac electric field. The emission peak occurred at about 600 nm from wires of average diameter of about 4 nm. The emission appears to result from band-to-band electron–hole recombination.

1. Introduction

Current interest in electrically driven nanoscale light emitters stems from their potential to be integrated with microelectronic devices such as lightwave components, for example, for optical interconnects. Among the various nanostructures, nanowires (NWs) and nanotubes (NTs) are well suited as light emitters since they are virtually defect-free, hence they allow for efficient radiative recombination. However, fabrication of light emitting devices composed of individual NWs and NTs is not simple since each one of these structures has to be individually connected to a set of electrodes for charge injection, as recently demonstrated [1, 2]. An alternate approach is to utilize an ensemble of NWs or NTs and capacitively couple them to an ac electric field, similar to ac thin film electroluminescent (ACTFEL) devices. Below we present our initial results demonstrating this approach to produce electroluminescence from silicon NWs.

Silicon has not been traditionally considered as a candidate for optical emission devices due to its indirect bandgap. However, electroluminescence (EL) has been previously demonstrated from amorphous silicon films [3] and more recently from porous silicon (PS) with solid-state contacts [4]. The source of the emission in PS has been attributed to the widening and formation of a direct bandgap resulting from quantum confinement in the silicon wires produced by anodization in aqueous or ethanoic HF solution [5]. Other possible sources of this light include defect states of SiO_2, interfacial interactions and surface states on the Si wires [6, 7]. PS is generally produced by anodization of Si in an HF containing solution, where the emission peak is dependent on the type of doping of the Si wafer and whether it was illuminated during anodization [8, 9]. In this investigation, rather than PS, we have employed Si in the form of NWs which were produced by catalytic decomposition of disilane on Au coated surfaces.

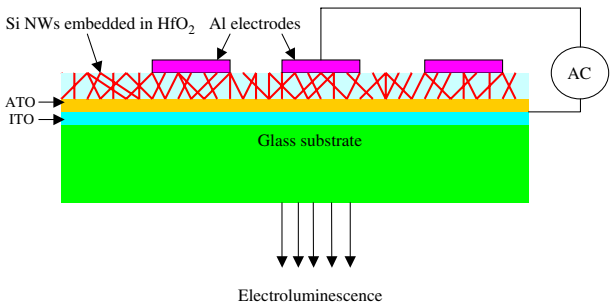

Figure 1. Schematic showing cross-sectional view of a Si nanowire electroluminescent device.
(This figure is in colour only in the electronic version)

2. Experimental details

The basic device design consisted of embedding NWs in a dielectric between two electrodes to form a capacitor, as illustrated in figure 1. To fabricate this device, NWs were first grown on glass substrates that had a pre-deposited 0.35 μm thick layer of indium–tin oxide (ITO), followed by a 0.26 μm thick layer of aluminium oxide/titanium oxide (ATO) composite, and finally a thin (1–2 nm) layer of gold. Both ITO and Au were deposited by sputtering and ATO via atomic layer deposition. The ITO layer formed the front (transparent) electrode. Silicon NWs were grown on the gold surface by thermal decomposition of disilane at 400 °C in a horizontal tube furnace described elsewhere [10]. One should note that in this device structure, since the nanowires are capacitively coupled, there is no contact resistance.

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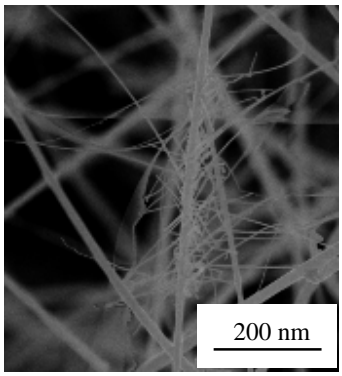

Figure 2. FESEM view of the Si nanowires.

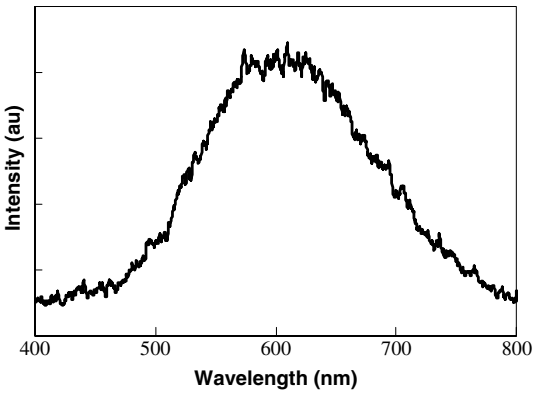

Figure 3. Electroluminescent spectrum of Si nanowires.

Si NWs of uniform diameter can be produced from disilane, where the diameter can be controlled via several parameters, including temperature. Also, finer Si NWs were observed to grow wrapped around the thicker nanowires. This is illustrated in figure 2 which is a field emission scanning electron micrograph (FESEM). The primary growth diameter of the NWs is about 20 nm, which are wrapped with about 3–4 nm diameter wires. It was difficult to precisely measure the diameters of these finer wires due to charging effects. Gold at the tips of the wider nanowires indicated a vapour–liquid–solid (VLS) growth mechanism [11]. However, at this time, we cannot explain the mechanism for the growth of the finer nanowires.

The next step involved deposition of an insulator to fill the gaps between the nanowires. This was achieved using atomic layer deposition (ALD) which produces a highly conformal film. The dielectric used for these devices was HfO_2 which has a relatively large dielectric constant ($\varepsilon_r \sim 21$), hence a large polarization vector. Dual metal precursors were utilized for deposition of HfO_2 at 170 °C, as described before [12]. The total thickness of this active layer was 1.5 μm. Following HfO_2 deposition, the substrates were annealed at 600 °C for 1 min in forming gas (10% H_2, 90% Ar) to densify the films. The final step involved deposition of 2 mm diameter, 0.5 μm thick Al electrodes, followed by a 450 °C, 1 min anneal in forming gas. Hence, a sandwich structure was produced that was composed of ITO as the front electrode, ATO, followed by a NW/HfO_2 active region, and finally Al discs as the back electrodes. The electroluminescence (EL) was viewed through the glass substrate. The EL produced by the Si NW device was coupled into an optical fibre and fed into a spectrometer (with a spectral range of 200–1000 nm) with a CCD array detector held at −120 °C.

3. Results

To observe the EL, the ITO and the Al back contacts were connected to a bipolar power source. At ±70 V and a pulse repetition rate of 3 kHz, one could observe the EL emission from these devices in a dark room. The glow of the EL, when viewed with a 10× microscope, was uniform across the 2 mm spot defined by the back electrode. The emission spectrum is shown in figure 3, where the peak occurs at about 600 nm (∼2 eV). There is no clearly defined peak, probably due to a size distribution of the NW diameters that leads to the optical broadening. This emission spectrum is similar to that recorded from PS produced with a cathode bias of −1.6 V [5].

4. Discussion

The mechanisms leading to EL emission from a conventional ACTFEL device are well established [13]. When a large electric field is applied across the electrodes, trapped carriers at the phosphor/insulator interface tunnel out and generate a displacement current within the phosphor. The impact excitation of the luminescent centres by the hot electrons leads to the optical emission. We propose a similar mechanism for the EL from the Si NW devices. Traps or interface states are expected at the Si NW/HfO_2 interface [14]. The applied ac field will induce tunnelling of electrons from these traps into the direct bandgap of the NWs, aided by quantum confinement. The electrical field is sufficiently large to accelerate these electrons to produce e/h pairs, which upon recombination produces the optical emission. Assuming band-edge to band-edge e/h recombination, the emission peak suggests a bandgap of about 2 eV. For a bandgap of 2 eV, both theoretical and experimental values of diameter to bandgap relationship of Si NWs indicate a NW diameter of about 2.2 nm [15, 16]. It appears that the finer (4 nm) diameter NWs are responsible for the light emission. Allowing for about a 1 nm thick layer of native oxide on the silicon nanowires, this would correspond to direct band-to-band recombination. There was no optical emission observed in dummy devices without the Si NWs. In the case of Si nanoparticles, it has been reported that photoluminescence peaks occur at about 800 nm, independent of the diameter. The source of this emission has been attributed to the existence of deep surface states resulting from dangling bonds on the surface of the Si nanoparticles [14]. As shown in figure 3, we do not observe any significant emission around 800 nm. X-ray excited optical luminescence studies of Si NWs has shown several emission bands in the visible region, where the band centred at 460 nm was attributed to triplet-to-ground-state transition in SiO_2 [17]. A weaker PL luminescence at 630 nm was attributed to interface defects between Si and SiO_2, and suboxide, as well as due to quantum confinement in Si crystallites (>2 nm) [17]. Hence, the EL observed in our devices is less likely to be originating from e/h recombination at the surface states or deep level states produced by defects due to Au (catalyst) impurity in the Si NWs. However, further

investigation is required to conclusively determine the source of EL we have reported.

5. Summary

The EL brightness and voltages reported above are for unoptimized devices. Both of these parameters can be significantly improved by growing only <5 nm wide Si NWs and refining the device structure. The device parameters such as the thickness of the NW plus the HfO_2 oxide layer and the ATO layer can be significantly reduced, which in turn would lower the operating voltage. Growth of more uniform diameter NWs would produce a narrower spectral distribution and their orientation perpendicular to the surface can significantly increase coupling to the electric field. For example, ZnO nanorods grown on A-plane sapphire would be ideal and could lead to laser emission. Our on-going work with intrinsically direct bandgap (ZnO, ZnSe) NWs show significantly brighter EL. These results will be published in the near future.

Acknowledgments

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