Low-temperature cathodoluminescence studies of GaAs nanowires in the SEM

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Abstract. We present data from cathodoluminescence (CL) studies of nanowires (NWs) grown from size-selected gold particles using metal-organic chemical vapour deposition. The NWs in this study all have a GaAs core and are surrounded by a shell of either AlGaAs or GaInP. The spatially and spectrally resolved CL studies were performed in a scanning electron microscope, in a temperature range of 4-300 K. The NWs were studied as-grown on their substrates in side view or transferred to Si substrates. We present data from single NWs, showing that the homogeneity of the emission is influenced by the growth conditions. A general observation is that the emission is much stronger, and more well-defined from the upper part of the NW. The shell emission varies in emission energy and spatial origin in an irregular fashion, probably linked to compositional variations in the shell.

1. Introduction

Over the past few decades, there has been a quest for the perfect system for fabricating high-quality quantum wires, or nanowires (NWs). Many approaches have been tried with varying successes, for examples see e.g. reference [1]. Semiconductor NWs seeded by metal particles have attracted much attention lately. In this study we present cathodoluminescence (CL) data from NWs grown by a technique introduced by Wagner [2] that has seen a revival over the past few years, due to improvements in the fabrication steps of the techniques [3].

The NWs in this study were grown with gold as the seed particles. The growth takes place at the interface between the metal particle and the semiconductor, taking its epitaxial information from the substrate. The growth conditions were chosen so that growth on the substrate surfaces is kinetically hindered and very limited, whereas material is precipitated at the interface between the seed particles and the semiconductor. The size of the seed particle determines the area of growth of the semiconductor. This results in the growth of pillars with a very high aspect ratio. With a random size of the particles, the diameter of the pillars is also random. However, with a well-controlled size of the particles, the resulting pillars have an identical diameter and identical height.
2. Experimental details
The gold particles used as seeds in this study were produced by an aerosol technique and size-selected, as described in detail elsewhere [4]. In this study, we concentrate on NWs grown from gold particles with a diameter of either 40 or 50 nm. Under the present growth conditions in our metal-organic chemical vapour deposition (MOCVD) chamber, the preferred NW growth direction is <111> [5] and we used (111)B-oriented substrates. The NWs were grown to a length of 1 - 6 μm. To avoid leaving the GaAs side-surfaces exposed, the NWs were covered by a layer of AlGaAs (or GaInP). This shell was grown at higher temperatures, where growth takes place without the aid of metal particles. However, there is still particle-mediated growth, giving an AlGaAs core at the top of the NW. This type of core-shell structure improves the light emission from the NWs by 2-3 orders of magnitude in resonant (excitation below the bandgap of the shell) photoluminescence studies [6]. During CL experiments, the emission has a tendency to fade permanently with time unless the shell is present. Depending on the sample, the shell thickness ranges from 20 nm to 55 nm in the present study.

The CL studies were performed in a dedicated scanning electron microscope (SEM) with a He cold stage. The NWs were studied as grown, still attached to the substrates, in side view, or broken off and transferred to Si substrates to avoid background emission from the GaAs substrate. The temperature was typically 6-10 K. The studies were performed at an acceleration voltage of 2.5-10 keV and a probe current of 10-100 pA. The choice of the acceleration voltage was based on two criteria, (1) The acceleration voltage must be high enough to penetrate the shell to the core and (2) The acceleration voltage must be low enough to keep most of the primary excitation in the NW.

3. Results
Figure 1 (a) shows an SEM image of a typical sample in the present study. There is a slight tapering towards the tip of the NWs, and at the bottom there is a “foot” caused by different growth facets developing around the base of the wire. Figure 1 (b) shows a typical low-temperature CL spectrum of several GaAs NWs. The 1.52 eV peak is related to excitonic emission in the GaAs core and the emission at 1.49 eV is related to carbon impurities in the core. In addition, emission in the range 1.42-1.50 eV can be caused by the mixed nature of the crystal structure, zincblende (ZB) and wurtzite (WZ) in combination with a type II band alignment, similar to what has been observed in InP NWs [7]. At the interface between the two crystal structures, spatially indirect transitions will result in an emission well below the energy of the lower bandgap. This is the ZB GaAs, as the WZ bandgap is higher [8]. If the segments are short, states might be quantized in the growth direction [9] and the energy of the transition is therefore higher than the minimum. It is worth pointing out that the cores of the present NWs are too large to exhibit any quantum confinement perpendicular to the growth direction.

Figure 2 shows a series of images from the same area of a typical sample, dominated by carbon-related emission. The CL images are recorded at different energies, as indicated on each image. The images have the intensity expanded to show the full intensity variation. For all images presented here, black means no emission. The main emission is much more prominent from the top of the NWs than the base, as shown in (b). The same pattern can be found for most of the NWs. Emission at lower energies, (c), is weaker and appears to originate further down the wire. Both these emissions come.
from the GaAs core. The emission from AlGaAs shell, (d), exhibits a spotty emission pattern without any trends. When varying the detection energy, the bright spots move around on the NWs in a random fashion, reflecting a local variation in the Al content. All the CL images of the GaAs emission lack emission from the top part of the NWs as the core in this part is AlGaAs, grown during the shell growth. The emission patterns are similar when the NWs are removed and transferred to non-emitting Si substrates.

![SEM image and CL images](image)

**Figure 2.** a) SEM image and the corresponding monochromatic CL images (b - d) of NWs with AlGaAs shells. The emission in (b) and (c) are from the GaAs core and (d) is from the AlGaAs shell. Especially (c) shows an increase in the intensity along the NWs. (d) shows a number of random emission spots along the NWs.

The increasing emission intensity towards the top is intriguing, and to understand this we have studied NWs of varying lengths in the same growth series. The emission from these NWs is dominated by excitonic emission. CL images from two samples in this series are shown in figure 3. These two sets of NWs are 3.5 μm and 5.5 μm long. They show similar trends in the emission pattern, where the first part is significantly weaker than the rest of the NW. In the case of the longer NWs, the upper micron even appears to be weaker in the image presented here. This effect can be reduced or enhanced by altering the scan speed and direction. One reason is that the long wires tend to vibrate under the electron beam leading to a lower excitation density. As the vibrations are more severe towards the tip, this leads to an apparent reduction in the emission intensity. Though not shown here, the emission from short NWs, 1-1.5 μm long, is very weak. This fits to the pattern of weak emission from the lower part of the NWs, irrespective of length.

![CL images](image)

**Figure 3.** CL images of a) 3.5 μm-long NWs and b) 5.5 μm-long NWs recorded at 1.51 eV. Both images show the same increase in the intensity from the substrate over the first 2-3 μm of the NWs. The longer NWs reach a constant intensity from about 3 μm.

Studying the increasing emission intensity along the NWs is an ongoing project. GaAs NWs can also be grown on other substrates than GaAs, like low-cost Si substrates. We have also used GaP substrates, usually in connection with GaInP shells. The GaInP shells are used as the strain induced by the shell can tune the emission energy over a large energy range. They are grown in a similar way, with the difference that the growth is initiated with a short GaP segment. These NWs exhibit a
different intensity pattern, figure 4. The main emission shows a decreasing intensity from base to tip, apart from the first 1-200 nm, which is GaP and therefore dark in the CL image of (b). In this case, the NWs are more tapered than the ones grown on GaAs substrates. The decreasing intensity is simply a volume effect where the diameter of the NW decreases and therefore the intensity decreases.

4. Discussion

The images presented here show that it is possible to study the emission from single NWs and to study variations along them as well as using spatially and spectrally resolved CL. Though not shown here, it is possible to record spot-mode spectra along the NWs in order to study variations in the emission. Line scans of NWs with AlGaAs shells confirm that the emission from the GaAs increases from base to tip. It also confirms the trend in figure 2 (b) and (c) that the peak position of the emission shifts towards higher energy along the NW. The intensity trend is the opposite of the growth on GaP substrates, where the intensity decreases along the NW. At the same time, the peak position is unchanged. The fact that the emission is strong already from the start shows that the effect is not intrinsic to the GaAs NW growth from gold particles but rather an effect of the substrate. We also have some preliminary indications that the surface preparation prior to growth can play a significant role in the emission intensity from the lower part of the NWs.

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