GaAs-based Nanowires Studied by Low-Temperature Cathodoluminescence

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Abstract. We present cathodoluminescence data of nanowires (NWs) grown using size-selected gold particles as seeds. The NWs have a GaAs core with a diameter of 50 nm and a length of several µm. The NWs in this study were generally covered with a shell of AlGaAs. With increasing growth temperature, the emission intensity increases significantly. From a variety of growth conditions, we conclude that the exposed sides of the NWs during growth play an important role in the emission intensity. The diffusion of carriers was studied by inserting a segment of GaInAs in GaAs NWs. By capping the NWs with an AlGaAs shell, we observe a tenfold increase in the diffusion length along the core.

1. Introduction
Over the past decades, there has been a quest to fabricate high-quality nanowires (NWs). These structures are interesting from a fundamental point of view. They also have potential for use in future devices [1]. Semiconductor NWs seeded by metal particles have attracted much attention lately. In this study we present cathodoluminescence (CL) data from NWs seeded by size selected gold aerosol particles [2]. One important figure of merit for NWs is the intensity of the emission. We have studied how the emission intensity is influenced by the growth temperature of the core. We have also studied how the diffusion of carriers along the core is influenced by the addition of a shell.

2. Experimental details
The GaAs NWs were grown using metal-organic vapour phase epitaxy (MOVPE), using 50 nm aerosol gold particles [2] to seed the growth on (111)B-oriented GaAs substrates [3]. The size of the particle determines the diameter or the resulting NWs, and the majority of the NWs grow in a <111>B direction. In order to study the effect of the growth temperature on the emission, a series of samples were grown in the temperature range of 380 to 500°C, in steps of 20°C [4]. The NWs were grown to a length of 3 - 4 µm, and for samples intended for studies of diffusion lengths (L_D), a short GaInAs segment was inserted. Most of the NWs were covered by an AlGaAs shell grown at a higher temperature (630°C), resulting in a shell thickness of about 20-30 nm, as determined by transmission electron microscopy. While growing the AlGaAs shell, some growth also takes place underneath the seed particles [9]. This results in the growth of a short AlGaAs NW core region at the top of the NWs.
Capping the NW cores with a higher band gap material (e.g. AlGaAs on GaAs) significantly improves the emission from the NWs.

The CL studies were performed in a scanning electron microscope (SEM) with a LHe cold stage. The emission was detected by a dedicated light-collection system [5, 6]. The NWs were studied in two modes: 1) as grown on the substrate in side view, and 2) broken off and transferred to Si substrates to avoid background emission from the GaAs substrate. The temperature was typically 6-10 K and monochromatic images as well as average spectra were recorded. An acceleration voltage of 5 keV and a probe current of 50 pA were used. Under these conditions, a spatial resolution along the NWs of about 50 nm can be achieved. We have used the inclusion of a low-bandgap segment (GaInAs, ≈5% In) in the core (GaAs) of a NW as a detector for diffusion into the segment. There is an intensity increase of the GaInAs emission when the electron beam is moved along the GaAs core towards the segment caused by diffusion from the core to the segment. Intensity profiles were extracted from monochromatic images and a single 1D exponential was fitted to each side of the intensity profile [7].

3. Result and Discussion

CL measurements were carried out on NWs grown under the different growth conditions described above. Spectra were recorded in side view from NWs grown at the different temperatures between 380 and 500°C. The scanned area typically contains 20-30 NWs. For each growth temperature, a reference spectrum was recorded by moving the sample to scan over that same area size on the side of the substrate instead. Two of the spectra (380 and 500°C) are shown in figure 1a. All spectra recorded for the NWs grown at different temperatures are dominated by a single peak at around 1.475 eV. The origin of the peak is not clear, but it appears to be related to twin boundaries in the nanowires. The main difference is that the intensity relative to the substrate increases with growth temperature of the core. The increase is about 30 times as the growth temperature is increased from 380 to 500°C. This increase has an exponential dependence on the growth temperature. This is shown in figure 1b, where the intensity ratio between the NW and the substrate emission is plotted on a logarithmic scale.

One source of the enhanced emission can be found in SEM images of the NWs. With increasing temperature, the NWs have a more tapered shape, as the higher temperature allows for some growth on the sides of the NWs during core growth, as can be seen in figure 2. This results in a slightly larger core diameter and thereby larger volume. We have observed that growth from larger-diameter particles leads to a higher emission intensity for NWs in the same growth run. However, this volume effect is not enough to explain the 30-fold increase in the intensity. Figure 2 also shows monochromatic CL images recorded at the peak of the NW emission. For the NWs grown at 380°C, the individual NWs are barely distinguishable in the image, whereas they are clearly visible at 480°C. For the low-temperature growth, the emission from the substrate is stronger than from the NWs. The ratio is reversed for the high temperature growth.

To further study the temperature effects, we have made a sample with a mixed-temperature core, started at 480°C and then switched to 380°C half way through the core growth. The emission from the entire core is as weak as for the 380°C core. We have also grown samples with a GaAs core and a...
GaAs shell, the latter grown at the normal shell growth temperature of 630°C. When covered by an additional AlGaAs shell, these NWs show weaker emission from the low-temperature core than from the corresponding NWs without the intermediate GaAs shell. However, the part of the GaAs core grown during the GaAs shell growth shows very strong emission intensity.

Figure 2. Two sets of images, a monochromatic CL image and the corresponding SEM image, from NWs grown at a) and b): 380°C and c) and d): 500°C.

The intensity of the emission depends strongly on competing non-radiative recombination. It is therefore essential to reduce the number of non-radiative recombination centers. For NWs, these are mainly associated with surface states and traps [8]. Non-radiative channels will reduce the total lifetime of the carriers and thereby also their diffusion length. We have investigated the effect of the shell on the diffusion length of carriers in the GaAs core, measured as described above on NWs transferred to Si substrates. The result is presented in figure 3. The monochromatic CL and corresponding SEM images are shown for a capped and an uncapped NW. The figure also shows two intensity traces along the NWs. By fitting exponentials to the intensity profiles (int=exp(-x/L_D)), it is possible to extract the diffusion lengths along the GaAs parts of the cores. In both cases, the diffusion length below the segment (left in the images and line traces) show similar values of about 100nm, which is much lower than expected for high-quality wires, e.g. about 3 µm for V-grooved wires [7]. Above the segment, there is a dramatic increase in the diffusion length for the capped core (>1µm) but there is almost no difference for the uncapped core.

Figure 3. a) SEM and b) CL image of an uncapped segment of GaInAs in a GaAs core. s) SEM and d) CL image of an AlGaAs-capped segment of GaInAs in a GaAs core. e) line traces of the intensity of the CL images of (b) and (c).

To explain the difference in the diffusion length below and above the segment for the capped core, we originally speculated that the carriers were lost to a radial GaInAs layer, a result of radial growth when growing the segment [9]. We observed weak emission at an even lower energy than the segment emission, which we attributed to a radial quantum well (RQW) with a higher In content than in the segment itself. However, this was on the edge of the range of the detector used so it was not clear. We have recently started to use a detector with a more suitable energy range and we have now been able to, not only confirm this but to show that the RQW emission is very strong. This is shown in figure 4 for NWs on the substrate, where the emission from the RQW is significantly stronger than the emission from the core and segment. Though not shown here, the emission pattern varies when the detection energy is varied, but it stays below the spatial location of the segment.
From the experimental results, the importance of the GaAs surface is clear. The lack of emission intensity is most likely related to the non-radiative recombination of traps on the GaAs surface [8]. It is evident from the measurements of the diffusion lengths that capping the wires with a passivating shell improves the diffusion length significantly. For the capped NWs, the improvement is related to a reduction in the surface recombination rate, related to surface traps [8], where fewer carriers are drained from the core. The same effect can be observed when the core is covered with a shell of lower bandgap material, a RQW. The difference is that the additional recombination path is radiative and can be observed, like in figure 4d. The draining of carriers by the RQW results in a similar diffusion length as for the uncapped core. As from the temperature dependence, the fact that low-temperature core growth on a high temperature-grown core results in emission intensity similar to the low-temperature core is significant. This means that the reduced emission intensity is probably a surface effect rather than a bulk effect, as the surface states become trapped and embedded in the structure. This leads us to believe that a higher growth temperature leads a lower density of surface states on the core, which in turn leads to stronger emission intensity.

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