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Carrier dynamics in InP nanopillar arrays fabricated by low-damage etching

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We present a comprehensive characterization of the optical quality of InP nanopillars (NPs) fabricated by a top down approach using micro-photoluminescence (µ-PL), time-resolved PL, and cathodoluminescence (CL). A lattice matched InGaAs layer provided beneath the 1 µm tall NPs functions as a “detector” in CL for monitoring carrier diffusion in InP NP. Carrier feeding to the InGaAs layer indicated by a double exponential PL decay is confirmed through CL mapping. Carrier lifetimes of over 1 ns and the appreciably long diffusion lengths (400–700 nm) in the InP NPs indicate very low surface damage making them attractive for optoelectronic applications.

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A tunable mode-locked Ti:sapphire laser (pulse temporal length \( \sim 100 \) fs, repetition rate 76 MHz, wavelength \( \sim 800 \) nm) with a spot diameter of 20–25 \( \mu \)m was used for excitation. The detected PL peak position was around 920 nm corresponding to the bandgap of epi-grown InP. As shown in Fig. 3, the measured PL decay curves show a double exponential behavior for the NPs on native substrate with the InGaAs layer. The PL decay for reference InP with the InGaAs layer shows a similar double exponential process (not shown here).

The measured TRPL decay curves were fitted using the following double exponential decay function:

\[
I(t) = I_0 + I_1 e^{-(t-t_0)/\tau_1} + I_2 e^{-(t-t_0)/\tau_2},
\]

where \( I(t) \) is the PL intensity at time \( t \) and \( I_0 \) at \( t_0 \). \( \tau_1 \) and \( \tau_2 \) are the PL lifetimes, for the two processes 1 and 2, respectively. \( I_1 \) and \( I_2 \) are constant coefficients. The fitted curves show very good agreement with measured TRPL data (Fig. 3). A fast decay time of 255 ps and a slow one of 1.15 ns were obtained for NPs on the InGaAs layer. The decay time of 1.15 ns is in the range of 500–775 nm, which is calculated from the relationship \( LD = \sqrt{D\tau} \), where the diffusion coefficients, \( D = 2.2–5 \) cm\(^2\)/s are used.

In order to confirm the draining of carriers to the underlying InGaAs and to determine the diffusion lengths in the InP segment of the NPs experimentally, CL measurements were performed using a dedicated SEM with a He cold stage. The acceleration voltage was 5 keV, the probe current 10–25 Pa, and the sample temperature 8 K. The NPs were investigated in side-view along the cleaved edge of the sample. The spatial resolution in CL imaging comes from the local excitation, where the detection has no spatial resolution. The spatial resolution is therefore limited by charge carrier diffusion in the sample under investigation and is used to determine ambipolar diffusion lengths from intensity profile. In the present structure, the lower bandgap material (InGaAs) can be used advantageously as a “detector” for the diffusing carriers in the InP NP. Fig. 4(a) shows a typical SEM image of an area with NPs together with monochromatic CL images in Fig. 4(b) for InP (\( \lambda = 876 \) nm) and in Fig. 4(c) for InGaAs (\( \lambda = 1490 \) nm). The InP emission intensity decreases towards base of the NP (Fig. 4(b)) while that of InGaAs decreases towards the top of the pillars (Fig. 4(c)), consistent with carrier transfer from the InP NP to the InGaAs layer.
Figure 5(a) shows the CL spectra of InP NPs and the InGaAs layer, and the peak positions correspond to their band-edge emission. The line scan, along the NP, obtained from the monochromatic ($\lambda = 876$ nm) CL image, is plotted on Fig. 5(b). The intensity is high at the pillar edge and drops exponentially towards the base. Conversely, the InGaAs intensity obtained from the monochromatic ($\lambda = 1490$ nm) CL image is the highest at the base and decreases exponentially towards the pillar edge. Due to the small diameter of the NPs, we can approximate the diffusion as one-dimensional, and the diffusion length can be determined by fitting a simple exponential to the intensity profile. The carrier diffusion length $L_D$ as determined by the single exponential fit made to the CL intensity profiles is $\approx 0.5 \mu m$ in InP (Figs. 5(b)–5(d)). Similar results were obtained from measurements made on several pillars. Here, we note that the change in the slope in CL intensity profiles (Figs. 5(b)–5(d)) around the InP NP/InGaAs interface region is due to direct excitation of the InGaAs layer. As the excitation spot gets close to the InP/InGaAs interface, a part of the generation volume is located inside the InGaAs layer. In other words, the break in the slope is when we go from pure diffusion in the InP NP to direct excitation of the InGaAs layer. Since the CL spectra of the InP epitaxial layer (peak at 880 nm) and InGaAs (peak around 1490 nm) are well separated, it is possible to combine line-scan data from the respective monochromatic CL images. Such a combined plot of the intensity profiles along the NP axis (Fig. 5(d)) clearly demonstrates carrier transfer into the InGaAs layer. The determined diffusion lengths are in good agreement with the value calculated from TRPL data. However, a rigorous determination of the diffusion length should include a full three-dimensional analysis.

In conclusion, we have performed a comprehensive analysis of the optical properties of InP NPs fabricated by a top-down approach. We demonstrated that these InP NPs have very good optical quality, comparable to as-grown epitaxial layers. The NPs show high PL intensities, PL line-widths as in as-grown layers even for single NPs, and appreciably long carrier lifetimes ($\approx 1$ ns) comparable to bulk values. The determined diffusion lengths in NPs using CL mapping are about $0.5 \mu m$. The appreciably long diffusion lengths make them attractive for optoelectronic devices including NW/NP solar cells both in axial and radial junction configurations. In addition, CL measurements directly confirm carrier transfer from the InP NPs to the InGaAs layer at their base, and demonstrate the use of a low-band gap layer as a detector to monitor carrier transport in InP NPs. For the specific InP NP/InGaAs structure investigated here, a carrier transfer rate of $\approx 250$ ps is determined by TPRL measurements.

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