Low temperature optical emission of WZ InAs NWs

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Abstract

Semiconductor nanowires (NWs) are attracting considerable attention due to their potential application in advanced nanophotonic devices. In addition, it has been shown that III-V NWs, such as InP, GaAs and InAs, can exhibit wurtzite (WZ) crystal structure in contrast to the cubic zincblende (ZB) phase of their bulk materials. The change in crystal structure alters the optical and electronic properties of the material and results in different fundamental physical parameters such as the band gap energy, exciton binding energy and phonon energies. As a consequence of the novelty brought by the NW growth, even for GaAs, which is among the most well-known materials, an ample controversy regarding the band gap energy of the WZ structure still exists. In the the case of InAs much less information is available. The known theoretical studies predict a WZ band gap 40 – 66 meV higher than the one of the ZB phase.[1,2] Trägårdh *et al*. have predicted a WZ band gap of 0.54eV by extrapolating fitted photocurrent measurements on $InAs_{1-x}P_x$ NWs[3] and Bao *et al*. observed a value of 0.52 eV in two-dimensional-like WZ structures.[4]

In this contribution, we have investigated the low temperature emission of WZ InAs NWs using PL spectroscopy. A detailed study of the power and temperature dependence of two InAs NW samples, grown at different temperatures, is presented.

The InAs NWs have been grown by the VLS method in a chemical beam epitaxy system using gold nanoparticles as catalysts on an (100) GaAs substrate. The growth temperatures were 420ºC and 450ºC for sample A and B, respectively. The structure of the NWs has been studied by transmission electron microscopy (TEM) and reveals a predominant WZ structure, grown along the [0001] direction, with presence of ZB stripes. The NWs of both samples have diameters in the range of 40 – 200 nm and lengths of 2 – 15 μm.

PL measurements were carried out on ensembles of NWs of sample A and B using a pulsed Ti-sapphire laser (pulse width of ~3 ps and repetition of ~80MHz) as exciting source syntonized at 760 nm and focused on a spot of approximately 1 mm^2. The sample was cooled with a cold-finger He cryostat with CaF2 optical windows, where the sample temperature was varied from 5 to 100 K.

In Fig. 1, the PL spectra from sample A [Fig. 1(a)] and sample B [Fig. 1(b)] measured for different excitation powers at 5K are presented. Two optical emission bands are observed in both samples. The low energy emission band (LEB) does not shift with increasing excitation power while we detect a pronounced blue-shift for the high energy emission band (HEB). In both samples, LEB dominates at low excitation power and HEB dominates for high power, suggesting a saturation effect which is common for an impurity-like recombination process at high excitation power, generally observed in bulk. The PL spectra at different positions on both samples were fitted with two Gaussian functions and their PL peak energies are depicted in Figs. 2(a) and (b). The HEB peak position blue-shifts approximately 15 meV as the excitation power raises from 10 to 500 meV. This large energy shift is a behavior usually observed in quantum wells (QWs) with type II band alignment which is attributed to the band bending induced by the carrier accumulation at the interfaces and the band filling effect. Similar blue shifts have been observed in InP[5] and GaAs[6] NWs containing WZ and ZB phases, where type II interfaces between these phases were present. In our InAs NWs, in fact, two phases along the wire axis are observed in TEM images forming QWs as depicted in the schematic diagram in Fig. 1(c). It has been predicted theoretically that InAs ZB/WZ superlattices have a type-II band alignment.[1] At low excitation powers, electrons in the ZB sections recombine with the holes in the WZ sections [transition 1 in Fig. 1(c)] leading to an emission below both, the ZB and WZ, band gap energies. When the excitation power is increased, state filling (transition 2) of electrons in the ZB section results in a blue-shift and broadening of the HEB [see Fig. 2(a)]. Thus, we attribute the HEB to the QW related emission. Our NWs present a diameter much larger than the effective Bohr radius and we assume that the lateral quantum confinement is negligible.

In Fig. 3(a) we present the temperature dependent PL spectra measured for a fixed intermediate excitation power (50mW) for sample B. In Fig. 3(b) we display the PL spectra for different excitation powers measured at 100K for sample B. The corresponding PL peak energies for the HEB as a function of temperature for a fixed intermediate excitation power (50mW) and as a function of different excitation powers measured at 100K are depicted in Figs. 3(c) and (d) for sample A (squares) and sample B (circles), respectively. A clear blue-shift with increasing temperature is observed. This behavior, which is opposite to the usual reduction of the band gap of semiconductors with increasing temperature, may be attributed to a temperature trigged band filling effect on the ZB phase, similar to that discussed for high excitation powers. In addition, rising the excitation power at 100K [see Fig. 3(b) and (d)] further blue-shifts the emission band. We associate this to the appearance of the band-to-band transition in the WZ phase. When the excitation power is increased at 100 K, this band becomes more evident and starts to dominate. This results in an overall blue-shift that is much larger than those observed in Fig. 2(a) attributed to the QW related emission. In fact, if we assume that at high temperatures the electrons in the QWs could be thermally excited to the WZ phase barrier, the recombination in the WZ phase [transition 3 in Fig. 1(c)] should increase while the QW related emission reduces. From the peak positions shown in Fig. 3(d), we estimate the fundamental gap of WZ InAs at 100K to be 0.453 ± 0.010 eV. Assuming that the temperature dependence of the band gap of InAs is the same for the WZ and ZB phases, the 5K band gap energy can be estimated as:

$$E_{WZ,5K} = E_{WZ,100K} + (E_{ZB,5K} - E_{ZB,100K}) = 0.453\text{meV} + 0.015\text{meV} = 0.468\text{meV}.$$

This value is 53meV higher than that of the ZB structure and is in very good agreement with the available theoretical works which predict a 40 – 66meV higher value for the WZ structure and it is close to the experimental data of 0.52meV and 0.54meV mentioned before.

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Figures

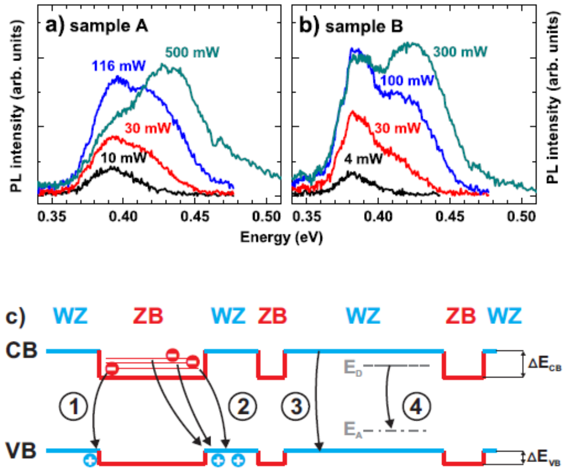

Figure 1 (top left): The PL spectra for different excitation powers measured at 5K from (a) sample A and (b) sample B, respectively. (c) Schematic diagram of the valence band (VB) and the conduction band (CB) of the WZ InAs NWs with ZB sections.

Figure 2 (down left): PL peak energies of the (a) high energy emission band (HEB) and (b) the low energy emission band (LEB) as a function of the excitation power for sample A (squares) and sample B (circles). Integrated PL intensities of (c) the HEB and (d) the LEB for both samples.

Figure 3 (down right): PL spectra from sample B measured at different temperatures. (b) PL spectra from sample B measured at 100K for different excitation powers. (c) PL peak energies of the HEB of sample A (squares) and sample B (circles) as a function of temperature. (d) PL peak energies at 100K as a function of excitation power.