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Novel Nanowires from Highly Mismatched Alloys for Optoelectronics

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Abstract

In this talk we review our recent results regarding optical properties and possible applications of novel nanowires (NWs) from highly mismatched alloys, such as GaNAs and GaBiAs. We show that their electronic structure can be easily tuned via alloying and lattice engineering, useful for their potential applications as nano-scale light sources within the infrared spectral region. We also demonstrate that short-range alloy fluctuations in these materials act as self-assembled quantum dots responsible for a three-dimensional quantum confinement of excitons, which can be utilized in single photon light sources.

1. Introduction

Owing to impressive advances in growth techniques, semiconductor nanowires (NWs) can now be fabricated from advanced materials that were previously available only in planar geometry [1]. One example of such new materials are highly mismatched III-V alloys, e.g. dilute nitrides and dilute bismites that are obtained from conventional III-V semiconductors by alloying with several percent of nitrogen or bismuth, respectively. These isoelectronic impurities cause strong perturbation of the electronic structure leading to a giant bowing in the bandgap energy. Therefore, GaNAs and GaBiAs are considered as promising materials for infrared light emitters.

In this talk we will show that fabricating these materials in the NW geometry provides the opportunity to combine their attractive physical properties with numerous advantages offered by the NW architecture.

2. Structures

All investigated NW structures were grown by molecular beam epitaxy on either (111) Si or (111) GaAs substrates. Both uniform GaBiAs, as well as core/shell and core/shell/shell structures with the GaAs core and the GaNAs or GaBiAs shell were studied. The former had wurtzite (WZ) crystal structure whereas the latter were crystallized in zinc blende (ZB).

3. Results and discussion

Effects of N and Bi incorporation on the bandgap energy in zinc blende NW structures

Alloying with N or Bi was found to cause a dramatic reduction of the bandgap energy, evident from a large red shift of the near-band-edge emission upon alloying [2, 3]. This is illustrated in Fig. 1 taking as an example GaAs/GaNAs core/shell NWs. Considering that the near-band-edge emission in GaNAs at low temperatures is caused by recombination of localized excitons (LE), the bandgap energy (E_g) in the alloy can be determined from the high energy cut-off of the LE band [1]. From Fig.1, an increase in the nitrogen content [N] by 0.5% caused a downshift of E_g by ~ 100 meV, illustrating flexibility of bandgap engineering via alloying in NWs from highly mismatched III-V alloys.

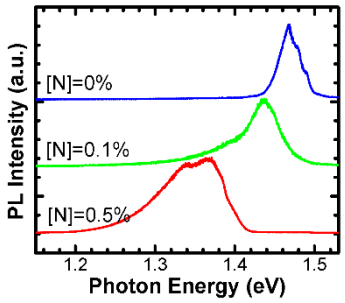

Fig. 1 Typical photoluminescence (PL) spectra measured at 4K from the GaAs/GaNAs core/shell NW arrays grown on a Si substrate.

Bandgap engineering via lattice engineering

One of important advantages of the NW geometry is the ability to tailor crystalline structure of the NWs. For example, though GaAs and related alloys can only be grown as ZB in bulk materials and epilayers, the related NWs can also be fabricated with the WZ crystal structure. Most recently [4], we have investigated how such changes in the crystal structure affect electronic structure of GaBiAs. By employing spatially resolved optical spectroscopies on individual NWs we showed that the bandgap energy of WZ GaBiAs is reduced as compared with parental GaAs. The polarization-resolved μ-PL and μ-PL excitation measurements showed that both the A and B valence subbands of the WZ GaAs NW are pushed upwards by anticrossing repulsion following incorporation of Bi atoms, while the symmetry order of the valence band states is kept unchanged under the current Bi compositions. Surprisingly, the shift was even larger for the C valence subband based on the performed resonant Raman measurements. The extraordinary modifications of the VB states are interpreted by expanding the valence band anticrossing model and assuming anisotropic hybridization energy in wurtzite GaBiAs NW. The model also predicts that the ordering of the A and B VB states can be switched in WZ GaBiAs alloys with larger Bi compositions, which could allow control of polarization direction of emitted and absorbed light in thin WZ NWs via

alloying. Furthermore, incorporation of Bi into GaAs significantly reduced the temperature sensitivity of bandgap in WZ GaBiAs NW. Our work, therefore, demonstrates that utilizing dilute bismide alloys provides new avenues for bandgap engineering and thus photonic engineering with NWs.

Lasing in GaNAs NWs

Utilizing the fact that an individual NW represents a naturally formed Fabry-Perot cavity, we have also demonstrated lasing from the GaNAs region of GaAs/GaNAs core/shell [5] and GaAs/GaNAs/GaAs core/shell/ shell NWs [6]. In both structures, increasing an excitation power (P_{exc}), caused lasing due to the GaNAs band-to-band transitions. The transition from spontaneous emission to lasing was accompanied by an 'S'-shape dependence of the PL intensity on P_{exc} and line narrowing- see Fig.2. By using rate equation analysis, a threshold gain, g_{th}, of 3300 cm^{-1} and a spontaneous emission coupling factor, β, of 0.045 were derived for the GaAs/GaNAs structure [4], whereas g_{th} = 4100–4800 cm^{-1} and β up to 0.8 were deduced for the core/shell/shell NW [6]. The performed simulations identify the HE21b cavity mode as the lowest threshold mode for lasing at 0.87 μm. The lasing mode changes to HE11a in the GaAs/GaNAs/GaAs core/shell/shell NWs with [N] ~ 2.5% that lase at 1 μm. This conclusion was supported by the polarization of the lasing line. From temperature dependence of the lasing emission, a high characteristic temperature, T_0, increases in the structures that have the outer passivating shell reaching 160(±10) K. Our results demonstrate a promising alternative route to achieve room-temperature NIR NW lasers thanks to the excellent alloy tunability and superior optical performance of dilute nitride materials.

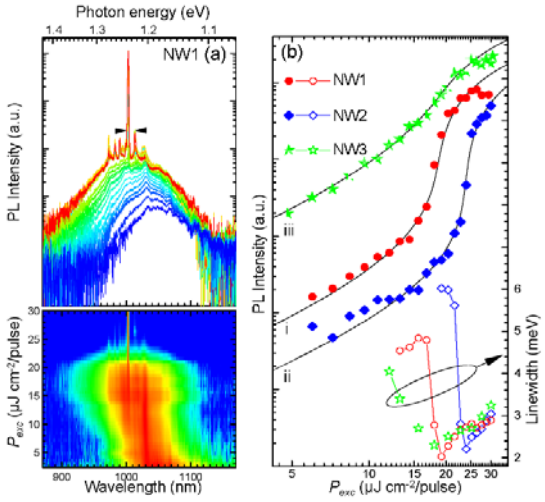

Fig. 2 (a) Power-dependent PL spectra of a representative single GaAs/GaNAs/GaAs core/shell/cap NW at 5 K. The lower panel is the corresponding mapping of the normalized PL intensity versus pump fluence. (b) Integrated PL intensity as a function of P_{exc} of the main lasing lines from several NWs. The filled symbols represent the experimental data whereas the solid lines are the best fit to the data based on the rate equation analysis. The linewidth of the PL signals versus P_{exc} are shown by the open symbols. From Ref. 6.

Formation of self-assembled quantum dots embedded in NWs from highly mismatched III-V alloys

We have found that incorporation of nitrogen in Ga(P)As NWs with both ZB and WZ structure causes local fluctuations in N composition, forming optically active and highly localized QD states inside the GaNAs shell acting as single photon emitters [7-9] – see Fig.3. The light emission from these localized states is found to be spectrally narrow (~50 - 100 μeV) and is highly polarized (up to 100 %) with the preferable polarization direction orthogonal to the NW axis, suggesting a preferential orientation of the localization potential. The electronic structure of these states is found to be controlled not only by confinement potential but also by strain induced by lattice mismatch between GaAs and GaNAs and local lattice distortion within short-range compositional fluctuations. Our results, therefore, show that alloying of GaAs with nitrogen is an efficient way to fabricate hybrid QD-NW structures with high crystalline and optical quality.

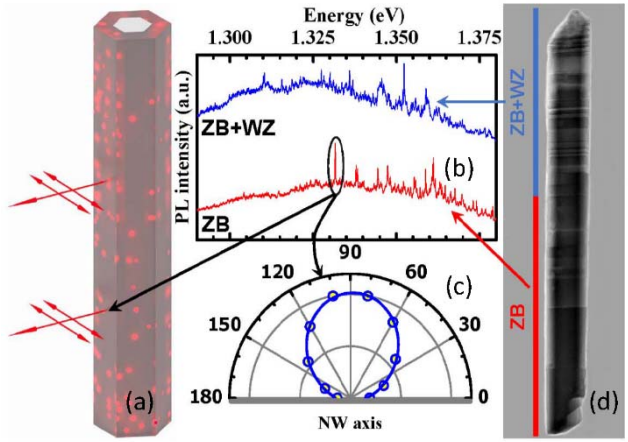

Fig. 3 (a) Schematic of the GaAs/GaNAs core/shell NW with QDs embedded in the GaNAs shell. (b) μ-PL spectra measured from the regions of the NW with different crystalline structure as shown in the TEM image (d). (c) A polar plot of the PL intensity of the QD emitter marked in (b). From Ref. 7

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