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Subcontractor Report

GaInNAs Structures Grown by MBE for High-Efficiency Solar Cells

Final Report
25 June 1999–24 August 2002

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NREL Technical Monitor: Richard Matson

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TABLE OF CONTENTS

GaInNAs Structures Grown by MBE for High-Efficiency Solar Cells

Abstract:

The focus of this work is to improve the quality of GaInNAs by advanced thin-film growth techniques, such as digital-alloy growth techniques and migration-enhanced epitaxy (MEE). The other focus is to further investigate the properties of such materials, which are potentially beneficial for high-efficiency, multi-junction solar cells. 400 nm-thick strain-compensated $Ga_{0.92}In_{0.08}As/GaN_{0.03}As_{0.97}$ short-period superlattices (SPSLs) are grown lattice-matched to GaAs substrates. The photoluminescence (PL) intensity of digital alloys is 3 times higher than that of random alloys at room temperature, and the improvement is even greater at low temperature, by a factor of about 12. The room-temperature PL intensity of the GaInNAs quantum well grown by the strained $InAs/GaN_{0.023}As$ SPSL growth mode is higher by a factor 5 as compare to the continuous growth mode. The SPSL growth method allows for independent adjustment of the In-to-Ga ratio without group III competition. MEE reduces the low-energy tail of PL, and PL peaks become more intense and sharper. The twin peaks photoluminescence of GaNAs grown on GaAs was observed at room temperature. The peaks splitting increase with increase in nitrogen alloy content. The strain-induced splitting of light-hole and heavy-hole bands of tensile strained GaNAs is proposed for explanation such behavior.

Project Objective:

To investigate novel materials for high-efficiency multi-junction solar cells (SC's).

Approach/Background:

To increase the conversion efficiency, tandem SC's, consisting of semiconductor SC's with different band gaps, are realized in monolithic or mechanically stacked design. Monolithic GaInP/GaAs tandem SC's, where GaInP has a larger bandgap than GaAs, has achieved record-setting efficiencies of 26% under the AM0 spectrum, and 30% at 500 suns terrestrial. If a SC with a bandgap smaller than that of GaAs can also be fabricated, the efficiency can be increased further.

Recently we and others have demonstrated that a new material, GaInNAs, can be grown lattice-matched to GaAs substrates and has a range of lower bandgaps than GaAs, in particular, 1.0 eV. We grow GaInNAs by gas-source molecular beam epitaxy using elemental Ga and In, thermally cracked arsine, and a RF plasma nitrogen radical beam source. This approach is expected to result in less carbon contamination as compared to metal-organic chemical vapor deposition. There are some materials issues, however. GaNAs and GaInNAs alloys tend to phase separate when the N composition is increased due to a large miscibility gap. Temperature-dependent photoluminescence (PL) and transmission electron microscopy study show that a concomitant presence of both In and N results in strain and/or composition fluctuations. In order to improve the quality of GaInNAs, we have investigated GaInNAs grown by migration-enhanced epitaxy (MEE), strained InAs/$GaN_{0.023}As$ and strain-compensated $Ga_{0.92}In_{0.08}As/GaN_{0.03}As_{0.97}$ short-period superlattices (SPSLs). In and N are separated in different layers.

Accomplishments:

Strain-compensated $Ga_{0.92}In_{0.08}As/GaN_{0.03}As_{0.97}$ short-period superlattices[1]

For high-efficiency multijunction solar cell applications the GaInNAs cell needs to have enough thickness to absorb the solar energy. In order to separate In and N and improve the GaInNAs quality, we grow $Ga_{0.92}In_{0.08}As/GaN_{0.03}As_{0.97}$ SPSLs instead of bulk $Ga_{0.96}In_{0.04}N_{0.015}As_{0.985}$. The layers in the SPSLs are strain-compensated, i.e., $GaN_{0.03}As_{0.97}$ is under tension and $Ga_{0.92}In_{0.08}As$ is under compression. Thus, thick $Ga_{0.92}In_{0.08}As/GaN_{0.03}As_{0.97}$ superlattices can be grown lattice-matched to GaAs substrates. Several SPSLs of $GaIn_{0.08}As/GaN_{0.03}As$ are grown with different periods from 16 Å to 100 Å. The number of periods is selected so as to keep the total thickness 0.4 µm constant. The zeroth order peak is closed to the GaAs substrate peak with a lattice mismatch less than 0.05%. Symmetric satellite peaks can be observed clearly.

The annealing temperature was investigated from 650°C to 900°C and optimized rapid thermal anneal condition for SPSLs is 700 °C and 10 sec in N_2 ambient. The PL intensity of SPSLs is much stronger than that of bulk layer. The PL intensity increases about 12 times at low temperature (10K) as shown in Fig. 1, and 3 times at room temperature.

Hall measurements show electron mobility is improved by a factor of almost two (240 vs. 130 $cm^2/V\cdot s$). The SL miniband effective mass can be theoretically calculated and is about half of the GaInNAs effect mass[2]. The improvement of electron mobility can be attributed to the smaller effective mass of the superlattice structure.

Photoconductive decay measurements show a longer carrier lifetime for the SL samples, 0.2 μsec vs. 0.1 μsec for the random alloy, which could be caused by charge separation due to the type-II band lineup of GaIn$_{0.08}$As/GaN$_{0.03}$As.

Strained InAs/GaN$_{0.023}$As$_{0.977}$ short-period superlattices[3]

For 1.3 micron GaInNAs lasers, It is indispensable to increase the In content (typically 30-35 %) and reduce the N content in the GaInNAs well layer. This results in a larger strain of over +1.5% in the quantum well (QW) active region and degraded QW quality. The growth temperature strongly affects the characteristics of GaInNAs. The optimal growth temperature range was found to be 440 - 450°C.

N incorporation forms N clusters in GaInNAs due to the large atomic size difference between In and N. The degradation of PL efficiency is originated from strong

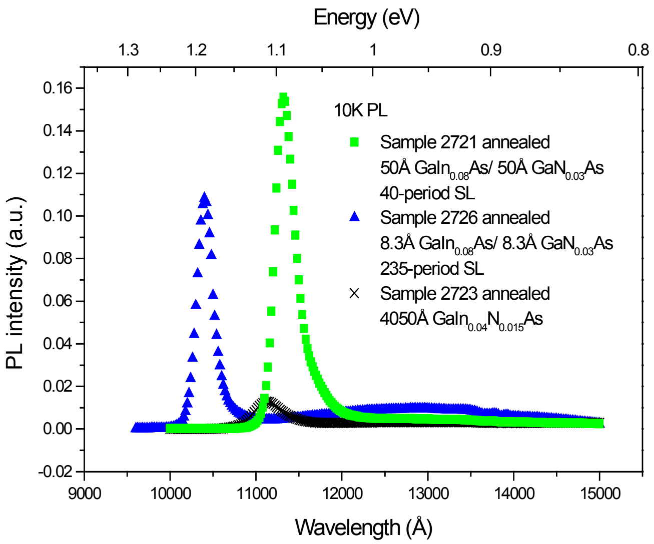

Fig. 1. Low temperature (10K) photoluminescence of superlattice samples and reference sample

carrier localization, which is due to the simultaneous presence of both In and N, and not solely of N. Spatial separation of In and N by the SPSL growth mode could be a way to improve the GaInNAs quality.

We grow $Ga_{0.67}In_{0.33}N_{0.015}As_{0.985}$ QWs using $(3Å\ InAs)_{10}/(6.2Å\ GaN_{0.023}As)_9$ SPSL mode (the inset of Fig. 2) and compare them with random-alloy $Ga_{0.67}In_{0.33}N_{0.015}As_{0.985}$ QWs. The SPSL growth method provides a simple way to tune the In and N mole fraction in QWs without changing the In and N beam flux. The PL intensity of the $Ga_{0.67}In_{0.33}N_{0.015}As_{0.985}$ QWs grown by the SPSL growth mode is higher by a factor 5 as compare to the continuous growth mode, as shown in Fig. 2

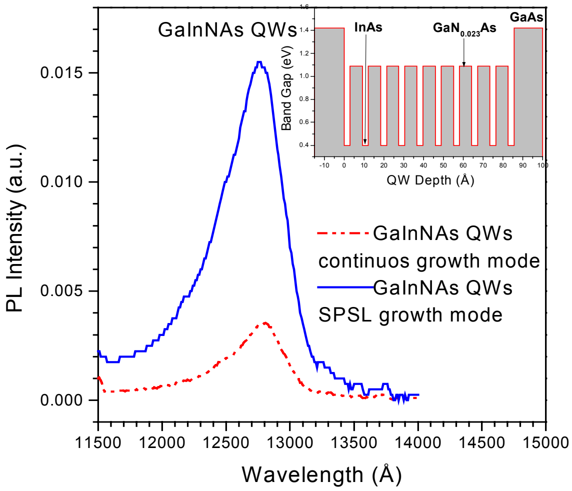

Fig. 2 Room temperature PL of $Ga_{0.67}In_{0.33}N_{0.015}As_{0.985}$ QWs grown by the SPSL mode and continuous mode. The inset is a detailed structure of $Ga_{0.67}In_{0.33}N_{0.015}As_{0.985}$ QWs grown by SPSL.

GaNAs grown by Migration-Enhanced Epitaxy[4]

We investigated the influence of MEE on the optical and structural properties of $GaN_{0.018}As_{0.982}$/ GaAs QWs. When group III atoms are deposited on a substrate surface in the absence of a group V overpressure, the migration length of the group III adatoms is increased. Two approaches are investigated. Alternating group III and group V flux onto the growing surface is called MEE, and introducing the group V flux alternatively while maintaining a constant group III flux is called modified MEE. Thus, low-temperature growth by MEE or modified MEE is expected to produce materials with comparable quality as those grown by molecular beam epitaxy (MBE) at higher temperature.

Each sample consists of a single 7-nm-thick $GaN_{0.018}As$ QW sandwiched between two 40-nm-thick GaAs barrier layers. The shutter sequence of each growth technique is schematically shown in the inset of Fig. 3. In order to increase surface migration of adatoms, a 2-second delay was introduced between consecutive shutter openings.

Fig. 3 shows low-temperature (8K) PL spectra of samples with QWs grown by MBE, MEE and modified MEE. The typical spectral shape of such emission is very asymmetric. The low-energy tail is reduced by modified MEE and even further by MEE. The QWs grown by MEE exhibit stronger PL intensity and smaller low-energy tail compared to the MBE-grown sample. The slope of the PL low-energy tail in a semilogarithmic scale can be used to estimate the localization potential E_o, as expected for a density-of-states tail caused by a fluctuation potential, $\rho(E) \propto e^{E/Eo}$. E_o is about 145 meV, 65 meV and 42 meV for MBE, MEE1 and MEE10 sample, respectively. The MEE samples exhibit clearly a reduced localization potential, indicating improvement in the GaNAs composition homogeneity.

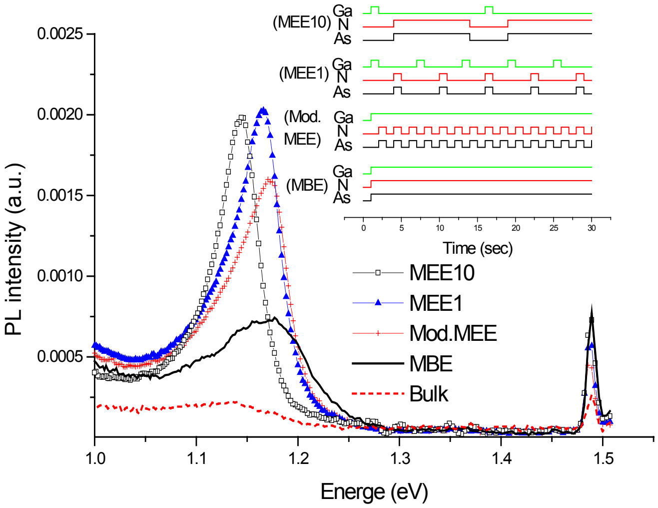

Fig. 3. Low-temperature (8K) PL spectra of QWs. PL measurements were taken under identical condition. The inset is a schematic representation of the different growth techniques used for the QW growth. (MEE1) is QWs grown by equal group III and V shutter opening time (1 second); (MEE10) is QWs grown by group V shutter opening time being 10 times longer than that of group III (10 seconds vs. 1 second). Bulk means a 100 nm-thick GaNAs sample grown by MBE.

Strain-induced valence band splitting in bulk GaNAs

Strained GaNAs layers were grown on (001) GaAs semi-insulated substrates. The PL spectra of GaNAs layers with varied nitrogen alloy content recorded at room temperature are shown in Fig. 4. The PL peak position shifts to longer wavelength side with increase in nitrogen alloy content. Each PL spectrum consists of two peaks and the energy distance between peaks increase with increase in nitrogen alloy content. This peaks splitting at room temperature become pronounced for GaNAs layer with nitrogen alloy concentration more than 1.5%. For lower concentration the homogeneous and inhomogeneous broadening of PL line screens this splitting. It is well known that nitrogen incorporation leads to inhomogeneous broadening of PL line due to nitrogen alloy content fluctuation. The typical inhomogeneous broadening of PL line for GaNAs alloy is about 30 meV. We have estimated the energy position of both peaks for samples with different nitrogen content. This dependence is shown in Fig. 5. It is clearly seen that the energy distance between the peaks of the PL spectra increase with an increase of nitrogen concentration, i.e. increase in tensile strain in the grown layers.

We follow Van de Walle's theory [5] in describing the dependence on strain of the valence- and conduction-band edges in III-V semiconductors. The effect of strain on energy levels can be decomposed into hydrostatic and shear contributions. Both contributions affect the valence band of zincblende semiconductors at Γ. The hydrostatic

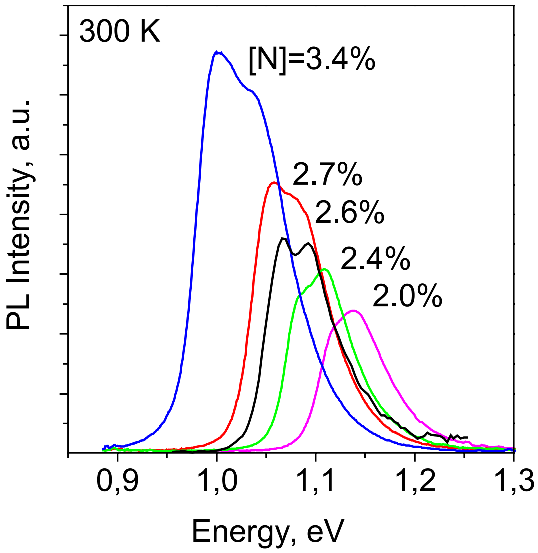

Fig. 4. Room temperature PL spectra of GaNAs layers with varied nitrogen alloy content grown on GaAs.

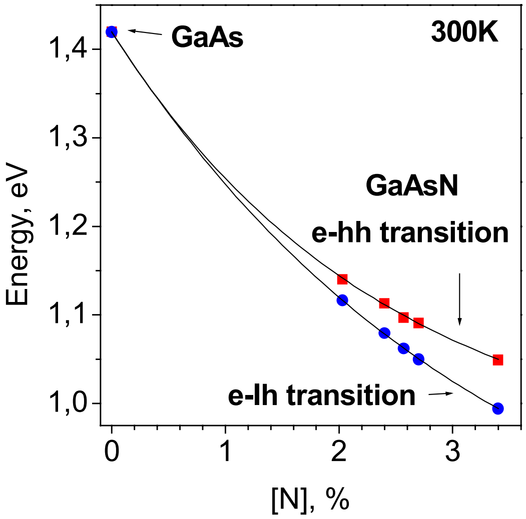

Fig. 5. The energy position of PL peaks for GaNAs with different nitrogen content as a function of nitrogen alloy content.

strain component leads to a shift of average valence-band energy, i.e., the average of the energies of the heavy-hole, light-hole, and spin-orbit split-off bands. The shear contribution couples to the spin-orbit interaction and leads to an additional splitting of the valence-band energies.

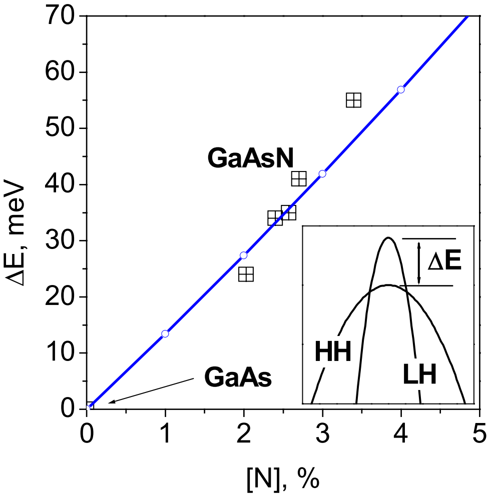

Fig. 6. The value of energy splitting of the heavy-hole and light-hole bands as a function of nitrogen alloy composition. The solid line shows the results of calculation. The squares show the experimental data, i.e. the energy distances between peaks of GaNAs PL spectra.

Using the above mentioned theory we have calculated the values of energy splitting, ΔE, between the heavy-hole and light-hole bands as a function of nitrogen alloy composition. The results of the calculations are shown in Fig. 6 (solid line). The energy differences between PL peaks for GaNAs with different nitrogen compositions, the experimental points, are also shown in Fig. 6 by the squares. The agreement with calculation is very good. Thus, we conclude that we have experimentally observed light- and heavy-hole transitions in GaNAs.

Other N-containing material systems

For completeness, we have also grown and investigated other N-containing materials, such as GaNP and GaInNP. We have found similar bandgap bowing as N in GaAs. Furthermore, a nitrogen concentration of 0.5% is the cross-over point from the indirect bandgap of GaP to direct bandgap of GaNP. Collaborations with NREL scientists and other groups in studying the properties of these materials as well as of GaNAs and GaInNAs have resulted in numerous publications, as listed below.

Conclusion

In order to improve the GaInNAs quality, we have demonstrated the growth of GaNAs ternaries and GaInNAs quaternaries using MEE and digital alloy technique, respectively, to separate In and N intentionally. 400 nm-thick strain-compensated $Ga_{0.92}In_{0.08}As/GaN_{0.03}As_{0.97}$ SPSLs are grown lattice-matched to GaAs substrates. The PL intensity of the digital alloys is 3 times higher than that of random alloys at room temperature, and the improvement is even greater at low temperature, by a factor of about 12. The room-temperature PL intensity of GaInNAs QWs grown by the strained $InAs/GaN_{0.023}As$ SPSL mode is higher by a factor 5 as compare to the continuous growth mode. The binary InAs allows for independent adjustment of the In to Ga ratio without group III competition. The In composition is easily adjusted by controlling the InAs growth time. MEE reduces the low-energy tail of PL. Two electron-hole transitions were observed in PL spectra for GaNAs alloys grown on GaAs substrate. Such behavior can be explained by the strain-induced splitting of light-hole and heavy-hole bands of tensile strained GaNAs alloy.

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13. ABSTRACT *(Maximum 200 words):* The focus of this work is to improve the quality of GaInNAs by advanced thin-film growth techniques, such as digital-alloy growth techniques and migration-enhanced epitaxy (MEE). The other focus is to further investigate the properties of such materials, which are potentially beneficial for high-efficiency, multijunction solar cells. 400-nm-thick strain-compensated $Ga_{0.92}In_{0.08}As/GaN_{0.03}As_{0.97}$ short-period superlattices (SPSLs) are grown lattice-matched to GaAs substrates. The photoluminescence (PL) intensity of digital alloys is 3 times higher than that of random alloys at room temperature, and the improvement is even greater at low temperature, by a factor of about 12. The room-temperature PL intensity of the GaInNAs quantum well grown by the strained $InAs/GaN_{0.023}As$ SPSL growth mode is higher by a factor 5 as compare to the continuous growth mode. The SPSL growth method allows for independent adjustment of the In-to-Ga ratio without group III competition. MEE reduces the low-energy tail of PL, and PL peaks become more intense and sharper. The twin peaks photoluminescence of GaNAs grown on GaAs was observed at room temperature. The peaks splitting increase with increase in nitrogen alloy content. The strain-induced splitting of light-hole and heavy-hole bands of tensile-strained GaNAs is proposed as an explanation of such behavior.

14. SUBJECT TERMS: PV; GaInNAs; digital-alloy; migration-enhanced epitaxy (MEE); solar cells; photoluminescence (PL); metal-organic chemical vapor deposition; strain-induced; splitting; short-period superlattice (SPSL)

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