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Near-infrared intersubband absorption in molecular-beam epitaxy-grown lattice-matched InAlN/GaN superlattices

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Strong near-infrared intersubband absorption is observed directly at room temperature in silicon-doped lattice-matched InAlN/GaN superlattices grown by molecular-beam epitaxy on GaN templates grown by hydride vapor-phase epitaxy. X-ray diffraction characterization of the heterostructures indicates excellent layer thickness uniformity and low interface roughness. For 2–4.5 nm quantum wells, the intersubband transition energies span the technologically relevant range between 2.3 and 2.9 μm. The experimental results are in good agreement with calculations of the transition energies using a conduction band offset of 1 eV and spontaneous polarization of 3 MV/cm. © 2009 American Institute of Physics. [DOI: 10.1063/1.3120551]

Extensive research in recent years on intersubband transitions in semiconductor heterostructures has resulted in numerous fundamental discoveries and technological device applications. Nitride-based materials are promising for intersubband light modulation, emission, and detection in the currently inaccessible 1.5–3 and 15–70 μm infrared ranges. The conduction band offset (CBO) between GaN and Al-containing alloys can exceed 1 eV, therefore allowing intersubband energy spacing in the near-infrared range. Intersubband absorption and luminescence around 1.5 μm from GaN/AlGaN heterostructures has already been reported. However, research efforts have been hampered by difficulties related to the structural quality of the materials. The material quality is limited in part by the lack of native nitride substrates. Heteroepitaxy of GaN on sapphire typically results in a high dislocation density that makes control of vertical charge transport difficult. Moreover, most of the intersubband studies reported in the literature employ high Al-content AlGaN alloys. The lattice mismatch between GaN and AlGaN limits the total thickness of material that can be grown before the structure relaxes by introducing additional dislocations and cracks. To eliminate the difficulties involved in growing strain compensated materials and theoretically calculating their band structure, it is preferable to develop a process for the growth of lattice-matched nitride superlattices possessing intersubband transitions in the near-infrared range.

In this letter, we focus on intersubband absorption in lattice-matched InAlN/GaN heterostructures. The large CBO and lack of a piezoelectric effect in the lattice-matched InGaN/GaN system makes it ideally suited for near-infrared optoelectronic applications. These lattice-matched nitride materials have been less investigated due to the challenges in growing high-quality InAlN. Photoinduced intersubband absorption in lattice-matched InAlN/GaN superlattices grown by metal-organic vapor phase epitaxy, as well as in molecular-beam epitaxy (MBE) grown strained InAlN/InGaN superlattices has been reported. Yet to date, no direct absorption data has been presented, nor has a detailed study of the absorption peak position and linewidth on quantum well (QW) thickness been performed. We present a detailed intersubband absorption study of lattice-matched InAlN/GaN superlattices grown by MBE on GaN templates grown by hydride vapor-phase epitaxy (HVPE). X-ray diffraction analysis suggests that our samples are of very high structural quality. The band structure of the superlattices was examined with Fourier-transform infrared spectroscopy. Strong intersubband absorption in the 430–520 meV energy range in 2–4.5-nm-wide QWs is reported.

The lattice-matched InAlN/GaN superlattices were grown by plasma-assisted MBE. To minimize threading dislocation density, the structures were grown on 50–100-μm-thick c-axis GaN templates grown by HVPE. These templates display smooth morphology and typical dislocation densities ~10^8 cm^-2. A systematic study of the MBE growth conditions of the ternary compound In_{x}Al_{1-x}N was presented elsewhere. This work showed that crack-free lattice-matched InAlN/GaN superlattices can be grown without changing the growth temperature for the alternating InAlN and GaN layers. Optimization of the growth of InAlN requires regulating the total metal flux to nitrogen flux ratio such that growth occurs near stoichiometric conditions. The superlattice samples discussed here consist of 20 GaN QWs with approximately lattice-matched In_{0.15}Al_{0.85}N barriers. The InAlN/GaN samples were grown at 540 °C as measured with optical pyrometry. Active nitrogen was supplied by a radio-frequency plasma source with power set to 400 W and a nitrogen flow rate of 0.5 SCCM (SCCM denotes standard cubic centimeters per minute at STP), resulting in a growth rate of approximately 140 nm/hr for InAlN. The GaN QWs were doped with Si to approximately 5 × 10^{19} cm^-3. For comparison of intersubband absorption, we also grew a 30 nm Al_{0.55}Ga_{0.45}N/2 nm GaN superlattice. This sample was grown at a higher temperature of 745 °C and as with the InAlN/GaN superlattices, the GaN wells were doped with Si at ~5 × 10^{19} cm^-3.

The structure of the MBE samples was characterized with a Panalytical X’Pert-MRD high-resolution x-ray diffractometer equipped with a 4-bounce Ge monochromator. Theoretical simulation of the diffraction data based on dy-
namical scattering theory with the commercial software package Panalytical X’PERT EPITAXY allows for an estimate of the well and barrier thicknesses, as well as the average InAlN composition. Figure 1 shows a typical x-ray diffraction spectrum for a 21-repeat superlattice consisting of 2.2 Å GaN QWs and 5.3 Å In$_{0.145}$Al$_{0.855}$N barriers. Except for the absolute intensity magnitude, the calculation of the x-ray pattern assuming sharp interfaces adequately reproduces the experimental data. The narrow satellite peaks and the visibility of thickness fringes (small secondary peaks) are indicative of low interface roughness and good thickness uniformity across the sample, as well as from well to well. The structural properties of the samples are comparable to the best reported for AlGaN/GaN superlattices grown by MBE.

For optical characterization, the samples were polished into multipass waveguides. In certain cases a metal layer was thermally evaporated on the surface to enhance intersubband absorption. The band structure was characterized at room temperature with a Thermo-Nicolet Fourier transform infrared spectrometer FTIR equipped with a beam condenser and wire grid polarizer. The infrared spectra were taken with the FTIR internal white light source and liquid-nitrogen cooled InSb detector in rapid scan. Infrared transmission of s- and p-polarized light was first normalized to the background, and then the ratio of the p- to s-polarizations was computed. Figure 2 shows the p/s normalized transmission for three QW widths. The exact position and full-width-at-half-maximum (FWHM) of the absorption features was extracted by fitting the spectra with a Lorentzian peak shape after subtracting a linear background. The results are detailed in Table I. For 2–4.5 nm QWs the intersubband absorption spans the energy range between 430 and 520 meV. The FWHM of the absorption peaks at the lowest energy is comparable with previously published values. We note, however, that the absorption profiles are broader than for transitions of corresponding energy in our AlGaN/GaN superlattice grown as a control sample. For comparison, Fig. 3 shows the intersubband absorption peak of a sample consisting of 30 2.2 nm GaN QWs with 4.0 nm Al$_{0.55}$Ga$_{0.45}$N barriers.

Calculations of the lowest intersubband transition energies were made within the envelope function approximation assuming different values for the CBO and spontaneous polarization field. Figure 4 shows schematically the profile of the CB for two adjacent 2.2 nm GaN QW surrounded by 2.2 nm InAlN barriers. For the two wider wells (3 and 4.4 nm), the transition energy is mainly determined by the triangular shape of the potential at the bottom of the well. Therefore, the spontaneous polarization field that sets the slope of the triangular well plays a more significant role in determining the transition energy than the CBO. Using this property, the spontaneous polarization field was estimated to be approximately 3 MV/cm. This value is consistent with experimental measurements of the photoluminescence. Moreover, for the narrowest well, the CBO plays an equally important role as

<table>
<thead>
<tr>
<th>QW width (nm)</th>
<th>Barrier width (nm)</th>
<th>$E_{\text{exp}}$ (meV)</th>
<th>FWHM (meV)</th>
<th>$E_{\text{calc}}$ (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.4</td>
<td>4.4</td>
<td>428</td>
<td>119</td>
<td>433</td>
</tr>
<tr>
<td>3</td>
<td>3</td>
<td>478</td>
<td>130</td>
<td>466</td>
</tr>
<tr>
<td>2.2</td>
<td>2.2</td>
<td>523</td>
<td>197</td>
<td>542 (miniband)</td>
</tr>
</tbody>
</table>

TABLE I. Experimental energies of the lowest intersubband transitions in a series of InAlN/GaN superlattices with the specified QW and barrier widths, and the corresponding calculated energies.
the spontaneous polarization field. Table I shows a comparison of the experimental transition energies with calculations assuming a CBO of 1 eV (Ref. 17) and a spontaneous polarization field of 3 MV/cm. For the two samples with wider wells, the barriers are thin enough so that the intersubband energy levels are well separated. For the sample with 2.2 nm wells, however, the barriers are thin enough (2.2 nm) so that the wave functions are delocalized, and therefore the transition energy listed in Table I is the separation between superlattice subbands calculated for a 6-well superlattice. The width of the top miniband for a 6-well superlattice was calculated to be 26 meV, while the energy difference between the two ground states (red and black curves) is 2 meV.

In summary, we have performed a detailed investigation of intersubband absorption in lattice-matched InAlN/GaN superlattices grown by MBE on thick GaN templates grown by HVPE. X-ray diffraction analysis of the samples indicates that the quality of the material is similar to the best reported In-free nitride heterostructures. Strong intersubband absorption features were observed directly at room temperature in the technologically important near-infrared range of (2.3–2.9 μm). The low limit of the range represents the shortest wavelength ever reported for the lattice-matched nitride heterostructures. The experimental results are in agreement with calculations of the transition energies using a CBO of 1 eV and a spontaneous polarization of 3 MV/cm. Our results prove the potential of the lattice-matched nitrides for quantum devices based on intersubband transitions and pave the way toward light emitters and detectors for spectroscopic applications based on this material system.

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