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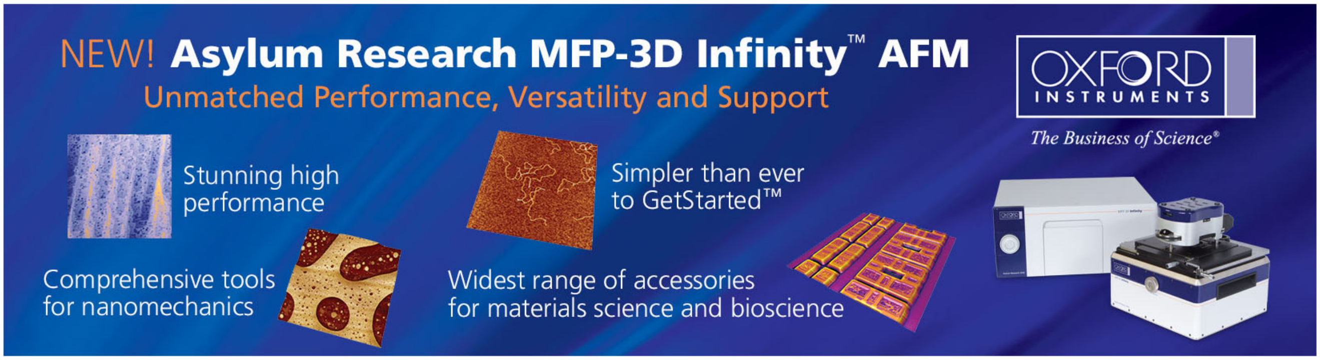

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Growth and deep ultraviolet picosecond time-resolved photoluminescence studies of AlN/GaN multiple quantum wells

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AlN/GaN multiple quantum wells (MQWs) with a well thickness of 26 Å have been grown by metal–organic chemical-vapor deposition. A specially designed photoluminescence (PL) spectroscopy system, which is capable of measuring picosecond time-resolved PL up to 6.2 eV, has been employed to probe the optical properties as well as the carrier transfer and decay dynamics in these MQWs. Optical transitions at 4.039 and 5.371 eV at $T=10$ K, resulting from the interband recombination between the electrons and holes in the $n=1$ and $n=2$ subbands in the wells, have been observed. The band-offset parameter for the AlN/GaN heterostructure has been obtained by comparing the experimental results with the calculations. Carrier dynamics including the relaxation of the electrons and holes from the $n=2$ and $n=1$ subband in the conduction and valence bands and the decay lifetimes of the interband transitions have also been measured and analyzed. Detailed subband structures for both the conduction and valence bands in the wells were determined. The implications of our findings on the potential applications of AlN/GaN quantum wells have been discussed. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1377317]

Significant progress has been made recently in III-nitride semiconductors.[1] Among the different structures and materials in III nitrides, AlN is the least studied and understood. With an energy gap of 6.2 eV at room temperature, AlN together with other III nitrides has many potential applications in optoelectronics, particularly in the UV spectral region. Most work for GaN/$Al_xGa_{1-x}N$ quantum wells (QWs) so far has focused on low-Al content $Al_xGa_{1-x}N$ barriers ($x\sim0.2$). Recently, the growth of GaN/$Al_xGa_{1-x}N$ QWs with x as large as 0.8 has been reported[2] and an effective confinement energy as large as 1.63 eV was observed for a 1-ML-thick QW. $Al_xGa_{1-x}N/Al_yGa_{1-y}N$ QWs ($x=0.11$) and QW light-emitting diode structures ($x=0.03$) have also been grown and studied.[3,4] However, the growth and optical properties of GaN/AlN QWs have not been reported. AlN/GaN QWs represent an unprecedented system with a bandgap difference as large as 2.8 eV, and thus offer many potential applications in optoelectronics, including UV detectors, modulators, infrared IR detectors, IR quantum-cascade (QC) lasers, etc.

In this letter, we report the growth and time-resolved photoluminescence (PL) studies of AlN/GaN multiple quantum wells (MQWs). PL emission lines from the interband recombination between electrons and holes in the $n=1$ and $n=2$ subbands in the wells were observed. The carrier transfer between different subbands as well as the decay lifetimes of the interband transitions have been measured and compared with the calculations. The band-offset parameter of the AlN/GaN heterostructure and the detailed subband structures for both the conduction and valence bands in the wells have been obtained by comparing the experimental results with the calculations.

The AlN/GaN MQWs studied here were grown by metal–organic chemical-vapor deposition (MOCVD). A 0.5 μm AlN epilayer was first deposited on a sapphire substrate with a 20 nm low-temperature AlN buffer layer, followed by five periods of AlN/GaN QWs with well and barrier thicknesses of 26 and 30 Å, respectively. The optimum growth temperature and pressure for the QWs were 1050 °C and 300 τ, respectively. In order to probe the emission properties and the carrier recombination dynamics in Al-rich AlGaN alloys, including pure AlN as well as AlN/GaN QWs, an unprecedented laser system was specifically designed to generate femtosecond (100 fs) tunable deep UV (195–200 nm) laser pulses with a 10 mW average power and 76 MHz repetition rate.[5] The system consists of a solid-state laser pumped Ti-

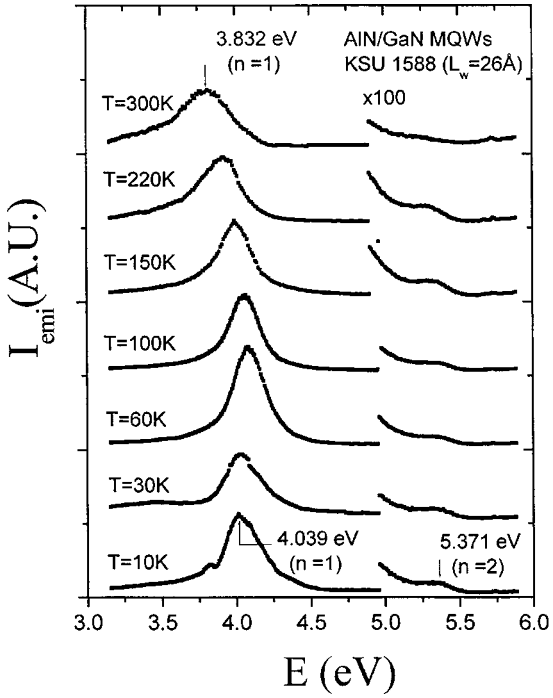

FIG. 1. PL spectra of AlN/GaN MQWs measured at different temperatures. The arrows indicate the peak positions of the PL spectra.

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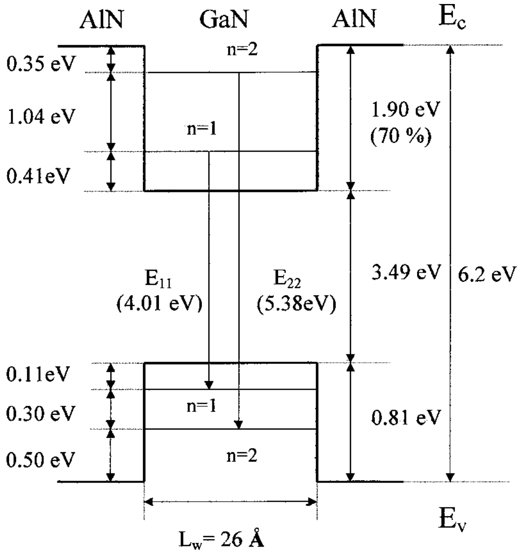

FIG. 2. Calculated subband alignment of AlN/GaN MQWs in the wells with the conduction-band-offset parameter, $\Delta E_c/\Delta E_g = 70\%$.

:sapphire laser and a specially designed frequency quadrupler. A 1.3 m monochromator together with a streak camera was used to record the time-resolved PL signal in the UV wavelength region with an overall time resolution of better than 10 ps.[5]

Figure 1 shows the PL spectra of an AlN/GaN MQW sample measured at different temperatures from 10 to 300 K. The arrows indicate the peak positions of the PL spectra. Two emission lines at 4.039 and 5.371 eV are observed at 10 K. The peak positions of both these two emission lines are redshifted with increasing temperature, as expected. We attribute both of the 4.039 and 5.371 eV emission lines to the interband transitions between the electrons and holes in the wells. The emission line observed at 4.039 eV corresponds to the optical transition of electrons and holes in the $n=1$ subbands in the wells, E_{11}. The emission line observed at 5.371 eV corresponds to the optical transition of the electrons and holes in the $n=2$ subbands, E_{22}. More than one subband is expected to exist for both electrons and holes in the wells due to the large band-gap difference between the AlN barrier and the GaN well. This assignment is also consistent with the fact that the emission intensity of the 4.039 line is much higher than that of the 5.371 eV line because the carrier populations (electrons and holes) are expected to be much higher in the first subbands (or the ground states). The interband transition, E_{12} (E_{21}), between electrons in the $n=1$ ($n=2$) subband and holes in the $n=2$ ($n=1$) subband, is not observable because E_{nm} ($n \neq m$) interband transitions in QWs are forbidden.[6]

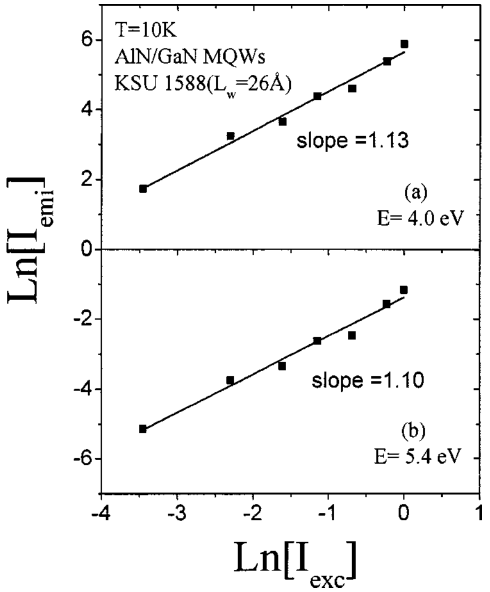

FIG. 3. Excitation intensity dependence of PL emission intensity, I_{exc} vs. I_{emi}, of AlN/GaN MQWs measured at $T=10$ K for the E_{11} and E_{22} transitions.

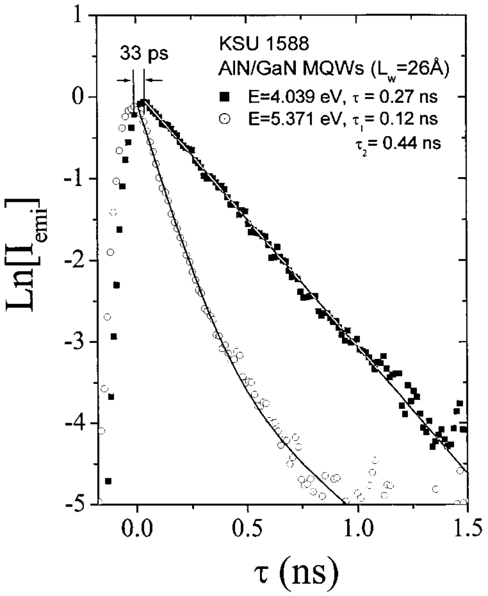

FIG. 4. PL temporal responses of the E_{11} and E_{22} interband optical transitions measured at $T=10$ K. The solid lines are the least-squares fit of data with single- and double-exponential decay formulas. A time delay of 33 ps for the PL intensity, reaching the maximum between the E_{11} and E_{22} interband transitions, is also observed and indicated in the figure.

A simple calculation was performed by solving a one-dimensional Schrödinger equation for electrons and holes in the QWs, neglecting the internal built-in fields (piezoelectric and polarization fields). We have used the following parameters for the calculation: $m_e = 0.2\ m_0$ and $m_h = 1.0$ for the electron and hole effective masses, and $E_g = 3.49$ and 6.20 eV for the energy gaps of GaN and AlN, respectively.[7,8] Table I lists the calculated optical transition energies of E_{11} and E_{22} for several different band-offset parameters. By comparing the calculated results in Table I with the experimentally measured emission energies of $E_{11} = 4.039$ eV and $E_{22} = 5.371$ eV, we see that a fairly good agreement between the calculation and experimental results is obtained when the conduction-band offset $\Delta E_c/\Delta E_g$ is adjusted to be around 70%. The detailed calculation results, including the band alignment with a conduction-band-offset parameter of 70% and the subband structure for the electrons and holes in the wells, are shown in Fig. 2. The effect of the internal built-in fields is expected to be larger on the E_{11} transition energy than on the E_{22} transition because the first subbands (E_1) of the electrons and holes are closer to the triangle regions in the conduction and valence bands in the wells. Hence, it is expected that the measurement and calculation results agree better for the E_{22} transition. The growth and PL studies for

TABLE I. Calculated E_{11} and E_{22} transition energies for several different band-offset parameters. $E_g(\mathrm{GaN})=3.49$ eV ($T=10$ K) and $E_g(\mathrm{AlN})=6.2$ eV ($T=10$ K).

		E_{11} (eV)	E_{22} (eV)
$T=10$ K (exp)		4.039	5.371
74%	$\Delta V_e=2.01$ eV	4.013	5.401
	$\Delta V_h=0.70$ eV		
72%	$\Delta V_e=1.95$ eV	4.011	5.390
	$\Delta V_h=0.76$ eV		
70%	$\Delta V_e=1.90$ eV	4.009	5.379
	$\Delta V_h=0.81$ eV		
68%	$\Delta V_e=1.84$ eV	4.007	5.364
	$\Delta V_h=0.87$ eV		
66%	$\Delta V_e=1.79$ eV	4.004	5.350
	$\Delta V_h=0.92$ eV		

AlN/GaN QWs have not been reported previously. Emission involving the excited states E_{22} has not been observed in AlGaN/GaN MQWs either. However, by extrapolating the conduction-band offset parameters from the GaN/AlGaN MQWs of varying Al content, a conduction-band offset of about 70% has been obtained,[9–12] which agrees well with our results obtained here.

The excitation intensity I_{exc} dependence of the PL emission intensity I_{emi} was also measured at 10 K for both the $n=1$ (4.039 eV) and $n=2$ (5.371 eV) transitions as shown in Fig. 3. The experimental results can be described very well by a power-law dependence of $I_{\mathrm{emi}} \propto I_{\mathrm{exc}}^m$. The fitted values of m for both emission lines at 4.039 and 5.371 eV are around 1.1, indicating an approximately linear dependence of the emission intensity with the excitation intensity for both of these two emission lines.

Time-resolved PL has been employed to probe the carrier dynamics in AlN/GaN MQWs. Figure 4 plots the PL temporal responses of the E_{11} and E_{22} transitions measured at the corresponding spectral peak positions at 10 K. The PL decay of the emission line at 4.039 eV ($n=1$) is clearly near a single exponential with a decay lifetime of about 0.27 ns. The PL decay profile of the E_{22} transition line at 5.371 eV is more complicated and is no longer a single exponential. Besides the radiative recombination, the photoexcited carriers relax thermally and optically from the $n=2$ to $n=1$ subband.[13] These processes couple together and lead to a double-exponential decay for the $n=2$ interband transition, as seen experimentally in Fig. 4. The solid line in Fig. 4 is a least-squares fit of the data for the E_{22} interband transition at 5.371 eV with a double-exponential decay formula. The fitted values of the two decay lifetimes are 0.12 and 0.44 ns. The longer decay lifetime of 0.44 ns is expected to be close to the radiative recombination lifetime of the electrons and holes in the $n=2$ subbands. Calculations also indicate that the radiative decay lifetime of the $n=2$ interband transition is almost twice as large as that of the $n=1$ interband transition, which further corroborates our interpretation.

It is also interesting to observe in Fig. 4 that there is a time delay of about 33 ps in the rise part of the PL temporal responses between the $n=1$ and $n=2$ interband transitions. We believe that this time delay is caused by the carrier relaxation from the $n=2$ to $n=1$ level. Under our experimental conditions, where photoexcitation at 6.2 eV is employed, most carriers are excited in the AlN barrier regions. These photoexcited carriers relax to the $n=2$ subbands first. Thus, the recombination between the electrons and holes in the $n=2$ subbands (with emission energy at 5.371 eV) occurs earlier than those in the $n=1$ states. It is important to realize that this type of relaxation process between different subbands in AlGaAs/GaAs QWs is very fast, typically, a few ps.[14] The relatively slow relaxation process of about 33 ps between the $n=2$ and $n=1$ subbands observed in the AlN/GaN QW here may be due to the much larger band offset involved for the electrons and holes as compared with the typical values of only tens of meV in the AlGaAs/GaAs system. However, the energy relaxation rate in the AlN/GaN QWs may be comparable to that in the AlGaAs/GaAs QWs.

With a large band-gap difference between AlN and GaN, many important applications for optoelectronic devices based on AlN/GaN QWs are conceivable. The interband transitions in the UV and deep UV regions have potential applications for UV detectors and emitters. The intersubband transitions in the AlN/GaN MQWs can be used for IR detectors and emitters as well as QC lasers.

In summary, AlN/GaN MQWs have been grown by MOCVD and studied by deep UV picosecond time-resolved photoluminescence. Emission lines due to the interband optical transitions of the electrons and holes in the $n=1$ and $n=2$ subbands have been observed. By comparing the experimental results with the calculations, a conduction-band-offset parameter of roughly 70% for the AlN/GaN heterostructures has been obtained. The carrier dynamics, including the decay lifetimes of the interband transitions of electrons and holes in the $n=1$ and $n=2$ subbands as well as the relaxation of the electrons and holes from the $n=2$ to $n=1$ subbands, have also been measured.

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