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G. Tamulaitis

K. Kazlauskas

S. Juršėnas

A. Žukauskas

M. A. Khan

See next page for additional authors

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G. Tamulaitis, K. Kazlauskas, S. Juršėnas, and A. Žukauskas[a)]
Institute of MSAR, Vilnius University, Naugarduko 24, 2006 Vilnius, Lithuania

M. A. Khan, J. W. Yang, J. Zhang, and G. Simin
Department of ECE, University of South Carolina, Columbia, South Carolina 29208

M. S. Shur
Department of ECSE and CIEEM, Rensselaer Polytechnic Institute, Troy, New York 12180

R. Gaska
Sensor Electronic Technology, Inc., 21 Cavalier Way, Latham, New York 12110

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We report on the spectral dynamics of the reflectivity, site-selectively excited photoluminescence, photoluminescence excitation, and time-resolved luminescence in quaternary AlInGaN epitaxial layers grown on GaN templates. The incorporation of a few percents of In into AlGaN causes significant smoothening of the band-bottom potential profile in AlInGaN layers owing to improved crystal quality. An abrupt optical bandgap indicates that a nearly lattice-matched AlInGaN/GaN heterostructure with large energy band offsets can be grown for high-efficiency light-emitting devices. © *2000 American Institute of Physics.* [S0003-6951(00)01440-6]

Recently, we demonstrated an energy band and lattice mismatch engineering approach using the quaternary AlInGaN material system.[1,2] The incorporation of In into AlGaN layers enabled us to significantly reduce the lattice constant of the alloy and, thus, to grow nearly lattice-matched AlInGaN/GaN heterostructures with large energy band offsets.[1] The introduction of AlInGaN barrier layers reduced the built-in strain and piezoelectric doping in AlInGaN/GaN heterostructures with a two-dimensional electron gas,[2] and significantly enhanced the light emission from AlInGaN/InGaN quantum-well structures and light-emitting diodes.[3]

The lattice match between AlInGaN and GaN layers is achieved at Al to In ratio close to 5 to 1, in good agreement with Vegard's law. However, the energy band offset in AlInGaN/GaN heterostructures extracted from the photoluminescence spectra is almost two times smaller than expected from the linear approximation of the energy gap in the quaternary AlInGaN alloys and the details of the energy band formation in the quaternary alloy are not clear. The incorporation of In into AlGaN requires reduced growth temperature in comparison with the growth of high crystalline quality AlGaN.[4] The growth at lower temperatures may lead to the disordering of the AlGaN sublattice and, thus, to the degradation of the material quality. On the other hand, indium is known to improve the crystal structure of III–*N* layers via isolectronic doping.[5,6] However, to date, the ordering effects and optical properties of the quaternary AlInGaN lack the systematic studies.

In this article, we report on a detail study of the optical bandgap in AlInGaN layers grown on GaN templates. We measured reflectivity, site-selectively excited photoluminescence (SSEPL), photoluminescence excitation (PLE), and time-resolved luminescence (TRL). Our results demonstrate that the incorporation of In into AlGaN layers results in a significant improvement of the material quality and optical properties of the quaternary AlInGaN alloy.

For this study, 200 nm thick $Al_{0.09}In_yGa_{0.91-y}N$ epitaxial layers were grown over a basal plane sapphire substrate using a low pressure metalorganic chemical vapor deposition. The growth temperature was 850 °C. A 100 nm GaN template was deposited prior to the deposition of AlInGaN. The Ga- and Al-precursor (TMGa and TMAl) flows were kept constant at an approximate ratio of 10. The incorporation of In was controlled by changing the flow of the In precursor, TMIn. The maximum flux of TMIn, F_{In}, was 10 μmol/min, which corresponded to the incorporation of approximately 2% of In. The detailed description of the growth conditions and sample characterization can be found in Ref. 1. The extracted values of In molar fraction did not account for possible corrections due to the biaxial and hydrostatic strain,[7] which may depend on the degree of relaxation[8] and on the defect density.

The reflectivity spectra were measured using a tungsten lamp. They exhibit an exciton-like structure. We used the spectra for the extraction of the exciton resonance energy by a conventional fitting procedure.[9] The extracted resonance energies are 3.677, 3.645, and 3.566 eV for an In-precursor flux of 0, 5, and 10 μmol/min, respectively. We also estimated the exciton energy in the GaN buffer to be 3.418 eV.

The SSEPL and PLE spectra were measured using a tunable organic-dye laser pumped by the fourth harmonic of a *Q*-switched YAG:Nd^{3+} laser (10 ns pulse duration). A set of dyes (buthyl-PBD, BMQ, p-terphenyl, and BM-terphenyl) allowed us to cover a wide spectral range from 3.30 to 3.80 eV. The constant density of the incident photon flux of 5×10^{22} $cm^{-2}s^{-1}$ was controlled using a calibrated photodiode gauge (Ophir/PD10). The SSEPL and PLE spectra were recorded using a 0.6 m double pass monochromator and a UV-enhanced photomultiplier (Hamamatsu R1463P). The

a)Electronic mail: arturas.zukauskas@ff.vu.lt

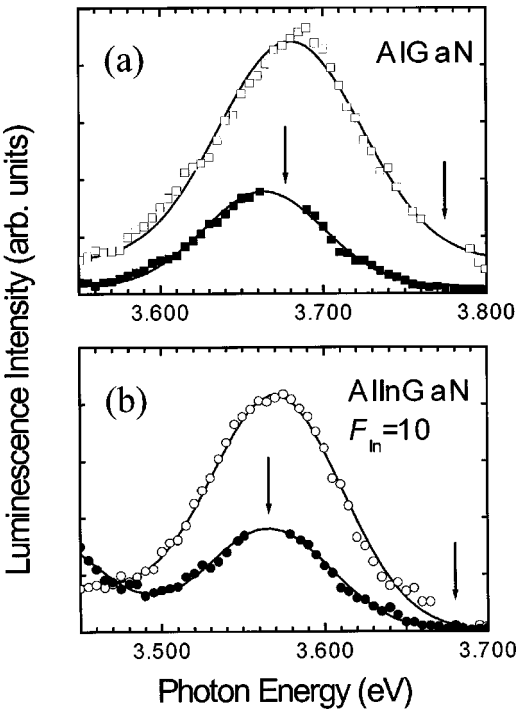

FIG. 1. Room-temperature luminescence spectra of AlGaN (a) and AlInGaN with $F_{In}=10$ μmol/min (b) layers. Open points, off-resonant excitation; filled points, resonant excitation at the exciton energy (arrows indicate the incident photon energy). Solid lines, Gaussian approximation.

luminescence spectra for the samples grown with indium fluxes $F_{In}=0$ and $F_{In}=10$ μmol/min are shown in Fig. 1. The data is presented for two incident pump energies. The solid dots in Fig. 1 are measured for the incident photon energies that coincide with the exciton energies extracted from the reflectivity spectra. The open dots correspond to the measurements for the incident photon energies that are approximately 100 meV above the exciton resonance. As seen from Fig. 1(a), the peak position of the resonantly excited luminescence band in AlGaN is about 13 meV below the exciton energy. As discussed below, this peak blueshifts toward the exciton position with an increase in the excitation photon energy. In contrast, the peak position of the luminescence band in AlInGaN samples (indium flows $F_{In}=5$ and $F_{In}=10$ μmol/min) is close to the exciton energy independently of the excitation photon energy.

Figure 2 shows the dependence of the luminescence band peak position on the incident photon energy (SSEPL data). In all investigated samples, a crossover from the anti-Stokes to Stokes luminescence is observed close to the exciton energy (indicated by arrows). The Stokes luminescence band in AlGaN layers (upper points) monotonously shifts towards the higher energies. Such behavior of the luminescence band is characteristic for the localized-state filling in disordered systems.[10,11] In contrast, no considerable shifting is observed in the quaternary AlInGaN samples. We attribute the observed changes in the SSEPL to the indium-induced reduction of the band-tail states in the AlGaN sublattice.

Our assumption about the reduction of the disorder in AlInGaN is also supported by the PLE data presented in Fig. 3. These PLE spectra show spectrally integrated intensity of the photoluminescence band in AlGaN and AlInGaN layers as a function of the incident photon energy. The integral PLE

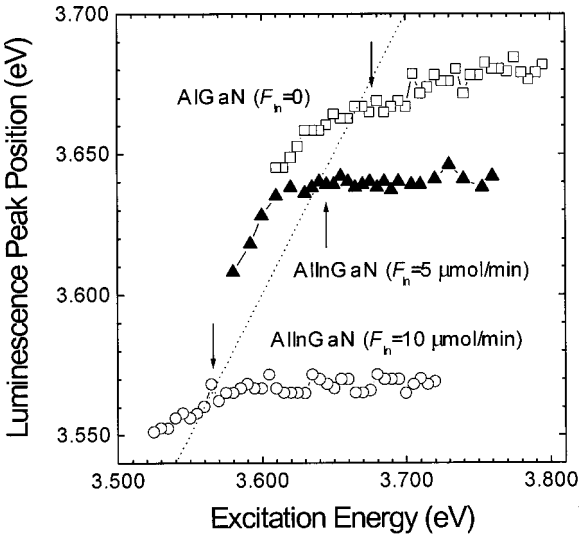

FIG. 2. Dependences of luminescence peak position on incident photon energy AlGaN ($F_{In}=0$) and AlInGaN ($F_{In}=5$ μmol/min and $F_{In}=10$ μmol/min). Dotted line separates the Stokes region (right) from the anti-Stokes region (left). Arrows indicate exciton positions.

spectra of the buffer GaN layer are also shown in Fig. 3 for comparison. As seen from the figure, an Urbach tail is clearly observed below the exciton energy in AlGaN layers. Above the exciton energy, the spectra exhibit a monotonous increase and saturation of the PL intensity. The energy region between the exciton energy and the saturation energy becomes smaller with incorporation of In. Note that in the buffer GaN layer, the saturation occurs immediately after passing the exciton energy (see Fig. 3).

We explain the difference between the PLE spectra of AlGaN and AlInGaN by the improved materials quality of the quaternary layers. The higher quality of AlInGaN results not only in the reduction of the band-tail states but also leads to improved carrier transport characteristics. When the absorption length of the incident light is larger than the nonequilibrium carrier diffusion length, an increase in the excitation energy results in a higher light absorption, and, thus, in a higher density of carriers and excited tail states. This, in turn, yields a higher photoluminescence intensity. At the excitation energies corresponding to the onset of the PL saturation, the absorption length becomes comparable to or less

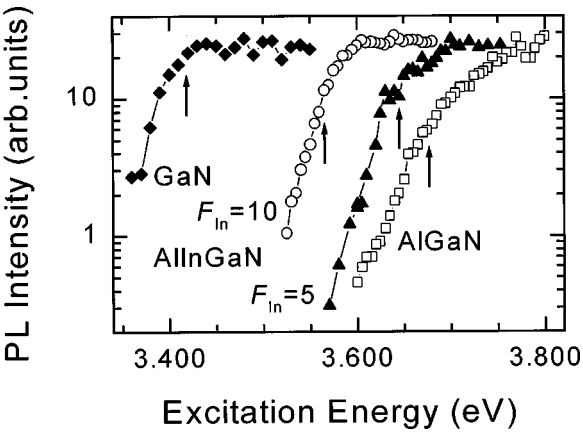

FIG. 3. Dependence of spectrally integrated luminescence intensity on incident photon energy in AlGaN, AlInGaN with $F_{In}=5$ μmol/min and $F_{In}=10$ μmol/min, and GaN buffer layer. Arrows indicate exciton energies.

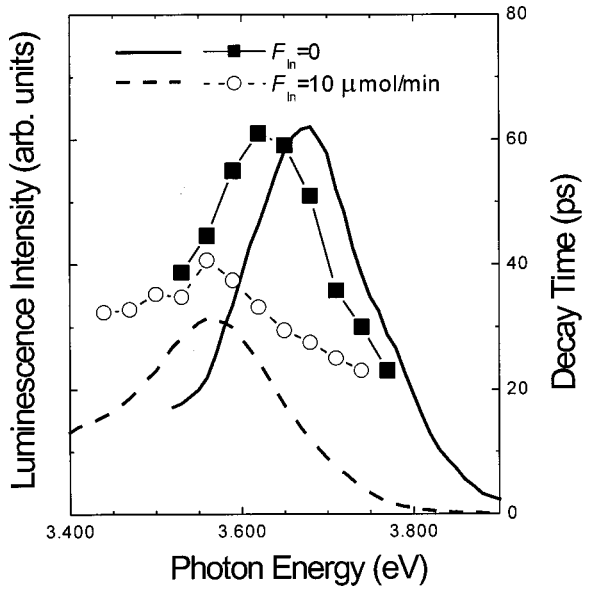

FIG. 4. Spectral distribution of luminescence decay time (points) and luminescence spectra (lines) in AlGaN and AlInGaN (F_{In}=10 μmol/min) layers.

than the diffusion length. In this regime, the nonequilibrium carrier density reaches its maximum value and stops growing. We should stress that the described behavior of PLE applies to the excitation with the constant number of incident photons.

In a partially disordered AlGaN, the reduced carrier diffusivity and, probably, damped exciton resonance prevent the PLE spectra from saturation. With the incorporation of indium (quaternary AlInGaN) the saturation is achieved earlier because of increased carrier diffusivity and reduced exciton damping. The diffusion length in a high-quality GaN buffer layer becomes larger than the absorption length at exciton energy. Therefore, we attribute the dynamics of the PLE spectra with increasing In molar fraction to (i) an increase in the carrier diffusivity associated with smoothing of the energy band potential profile, and (ii) the intensification of the band gap resonance.

The improvement in the crystal quality of the quaternary AlInGaN with increase of In content was also supported by the TRL results. For TRL measurements, we used a frequency-quadrupled and mode-locked Nd^{3+}:YAG laser (photon energy 4.66 eV, pulse duration 20 ps). The temporal resolution of the measurements close to 20 ps was achieved using a toluene optical Kerr shutter. Figure 4 shows the spectral dependence of the luminescence decay time in AlGaN and quaternary AlInGaN layers. The decay time, which was extracted from the exponential kinetics of the luminescence intensity in the late stage of the relaxation, is collated with the corresponding luminescence spectra. As seen from the figure, a shift of the decay-time spectra towards lower energies by more than 50 meV is observed in AlGaN. We attribute this shift to the carrier localization effect in the band-tail states of a partially disordered crystal.[10,12] With incorporation of In in quaternary AlInGaN layers, the spectral distribution of the decay time flattens, the peak decay time decreases, and the shift disappears. These TRL features point to a transition from a localized- to delocalized-carrier regime.

In conclusion, we have demonstrated that incorporation of a few percents of indium into AlGaN significantly changes the band-edge optical properties of the layers. AlGaN layers under study are partially disordered because (i) the layers were grown at reduced temperature and (ii) they are partially relaxed (since estimated critical thickness of the fully strained AlGaN with 10% of Al is less than 200 nm). The chaotic potential distribution in AlGaN causes the blueshift of the luminescence band with increased incident photon energy (filling of the band-tail states), the occurrence of an unsaturated region in the PLE spectra (reduced carrier diffusivity due to the potential ''roughness''), and the redshift of the luminescence decay-time spectral distribution with respect to the luminescence band (carrier localization). The SSEPL, PLE, and TRL data indicate that the initial partial disorder, which is characteristic of the AlGaN sublattice disappears with the incorporation of In. The improved structure of the alloy might be related to isoelectronic doping similar to that observed in GaN.[5,6] An abrupt band gap is formed in AlInGaN with approximately 2% of In, which corresponds to a nearly lattice-matched growth of $Al_{0.09}In_yGa_{0.91-y}N$ on GaN. These results demonstrate a potential of energy gap and lattice engineering using the quaternary material system for the development of electronic and high-efficiency light-emitting devices.

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