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Transient photoreflectance of AlInN/GaN heterostructures

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Time-resolved photoreflectance (PR) in AlInN/GaN heterostructures was applied to study carrier dynamics at energies extending from the uniform AlInN alloy band gap to the band gap of GaN. PR at the AlInN band gap has been found to have subpicosecond decay. Such ultrafast carrier relaxation from the extended to the sub-band edge states implies that the localization sites are small and dense, most probably originating from the In-rich clusters. At energies below the AlInN band gap, a complicated energy dependence of the PR signal is attributed to the properties of the localized states and to the modulation of the interface electric field by photoexcitation. Copyright 2012 Author(s). This article is distributed under a Creative Commons Attribution 3.0 Unported License.[http://dx.doi.org/10.1063/1.4768670]

Al1-xInxN is the least researched of ternary nitride alloy semiconductors. In comparison with AlGaN and InGaN, localization potentials in AlInN (characterized by the Stokes shift) are much larger. This property may have a big impact on electronic and optical applications of AlInN. In AlGaN, the energy difference for optical transitions between the extended and the localized states is 30 to 80 meV,1 in InGaN – 20 to 100 meV;2,3 In AlInN, for application-relevant In fraction around 17% (lattice matched to GaN), the Stokes shift can be as large as 1 eV.4–6 Several mechanisms might be responsible for the shift. First, it may be induced by large and random alloy composition fluctuations affecting potentials in the conduction and the valence bands. (In other ternary nitrides, e.g. InGaN, the energy minima related to the composition fluctuations trap carriers and prevent them from nonradiative recombination.)2,7 The alloy composition variations in AlInN might be related to structural defects. A study by Mouti et al.8 revealed that composition variations generated by strain fields occur around dislocations. Dislocation-related In-rich regions could trap carriers and aid the nonradiative recombination via point defects typically agglomerated around dislocations.9 These regions might also contribute to an efficient transport along dislocation-related high conductivity channels.10–12 Second, Gorczyca et al.13 showed that localized (sub-band edge) states might be related to the cation ordering around nitrogen atoms. For four In atoms surrounding a N atom (a so-called In cluster), the valence band minimum is about 1 eV higher than for a uniform alloy in which (taking Al0.75In0.25N as an example) three Al and one In atoms surround a N atom. The In clustering affects only the valence band and not the conduction band, which originates from the Al atomic orbitals. Therefore, the variations of the alloy composition and In clustering should affect the spatial uniformity of the conduction band triangular well at the interface of AlInN/GaN heterostructures in a different way. The height of the well depends on the conduction band offset, and the potential profile – on the spontaneous polarization difference between AlInN and GaN. While spatial AlInN composition variations affect both of these factors, nanoscale In clustering should only change the polarization. Since the triangular well serves as a channel in AlInN/GaN high electron density...
mobility transistors, the nature of the sub-band edge states might have a significant influence on device performance, even with the presence of an AlN interlayer.

In this work, sub-band edge states in AlInN have been explored via their influence on photoexcited carrier dynamics. This has been done studying the time-resolved photoreflectance (PR) from AlInN/GaN heterostructures. Previously, carrier dynamics in AlInN has been investigated only in the lowest sub-band edge states by time-resolved photoluminescence (TRPL). By varying the pump and probe photon energy from the uniform AlInN alloy band gap all the way to the band gap of GaN, we have been able to assess the carrier dynamics in the extended states and in the entire spectrum of the sub-band edge states.

The study was performed on two undoped AlInN/GaN heterostructures. The first one contained a 95 nm thick Al0.86In0.14N layer and the second one - a 80 nm thick Al0.82In0.18N layer. In both samples, the AlInN layers were grown on a 2.5 \( \mu \)m undoped GaN layer. The samples were grown on c plane sapphire using migration-enhanced metal-organic chemical vapor deposition (MEMOCVD). The electrical and structural data for the structures can be found in Ref. 15. An Al0.86In0.14N/GaN structure with a 10 nm thick AlInN layer and a GaN epitaxial layer were used for comparison.

Degenerate PR was measured using a wavelength tunable (680–1080 nm) frequency tripled fs Ti:sapphire laser (150 fs pulse duration, 80 MHz repetition rate) and a standard lock-in technique. The typical average pump power density was about 300 W/cm\(^2\), which corresponds to the photoexcited carrier density of \( \sim 5 \times 10^{17} \text{ cm}^{-3} \). The probe power density was about 0.05 of that of the pump. At all wavelengths, the PR signal was proportional to the pump power density with no saturation effects observed. The measurements were performed at room temperature.

Figure 1 shows PL spectra of the two samples. The spectra consist of a sharp GaN and a broad AlInN peaks. For the Al0.82In0.18N/GaN structure, these peaks overlap; oscillations in the AlInN peak are due to the Fabry-Perot interference. The inset shows the PL excitation (PLE) spectrum for Al0.86In0.14N. The PLE peak corresponds to the uniform alloy band gap energy for the studied layer, in good agreement with the value determined by spectroscopic ellipsometry. PL is generated by transitions involving the sub-band edge states. The Stokes shift, described as the difference between the PL peak and the band gap, is about 0.8 eV for both samples. The large AlInN peak width indicates that the sub-band edge states in AlInN have a broad spectrum. Different patterns of In clustering that involve atoms beyond the nearest neighbors might account for this effect.

Since the TRPL produced no signal at the uniform alloy band gap, a pump and probe experiment in the reflection geometry was applied to study the carrier dynamics in the extended states. The interpretation of the time-resolved PR results, however, is less straightforward, since reflectance is an integral parameter. In general, the change of the probe reflectance, \( \Delta R \), depends on the change of both the real and imaginary parts of the complex refraction index \( n' = n + ik \). For excitation in
FIG. 2. PR transients for Al$_{0.86}$In$_{0.14}$N/GaN structure with the 95 nm thick AlInN layer.

FIG. 3. PR decay times (a) and signal amplitudes (b) measured for the 300 W/cm$^2$ average pump power density for AlInN/GaN heterostructures. The arrows indicate uniform alloy AlInN band gaps.

the band gap region, the contribution of $n$ is dominant. The change of the refractive index is determined by the change of the extinction coefficient $k$ through the Kramers-Kronig relation. A decrease in $k$ due to absorption bleaching (state filling) increases the refractive index at energies exceeding the band gap, with a diminishing weight as the energy difference between the filled and probed states becomes larger. The reflection is affected by the change of the electron and hole distribution functions ($f_e$ and $f_h$), since $\Delta k \propto -(f_e + f_h)$. For the photoexcited carrier densities used in this study, the change of the reflection due to the band gap renormalization may be neglected. At the photon energies around the band gap, free carrier absorption may be ignored as well. Thus, in a bulk semiconductor, the $\Delta R$ signal is primarily determined by filling of electron and hole states.

Qualitatively, the time-resolved PR of both heterostructures follows the same pattern. Fig. 2 shows selected transients for the 14% In sample. The PR signal experiences a single- or a bi-exponential decay. Fig. 3 shows the decay times (the shorter components in the case of the bi-exponential decay) and the transient amplitudes for the same photoexcited carrier density as functions of energy. In the investigated photon energy range, one can distinguish several regions. At the energies above the band gap, the PR decay is composed of two components with a prevailing shorter decay time of 0.3–0.5 ps. At the band gap, the fast decay time is 0.6–0.9 ps and the slow decay time is 30 to 50 ps with the weight of the fast component being about several times larger. At energies below the uniform alloy band gap, the decay time increases until it saturates at about...
FIG. 4. Schematic diagram of an AlInN/GaN heterostructure. The short dashes with dots close to the valence band edge denote the sub-band edge states in the In clusters, the longer dotted dashes – the localized states related to AlInN composition fluctuations. Process A stands for the interband transitions over the uniform alloy band gap. Process B – for the electron absorption between the localized valence and conduction bands states, and between the In-cluster related valence band and extended conduction band states. Process C indicates the electron transitions from the In-cluster related valence band states to the conduction band with the subsequent electron transfer to the well or into the bulk of the AlInN layer. Process D stands for the interband absorption in GaN.

40 to 50 ps. At still lower photon energies, the transients abruptly change sign, and the decay time drastically decreases. In the transition region, one can observe a superposition of the fast negative and a slow positive response (4.18 eV transient, Fig. 2). Subsequently, the time constant of the negative transient increases, and, close to the GaN band gap, the transient becomes positive again. Along with the sign change, a strong increase of the \( \Delta R \) amplitude is observed as well.

The ultrafast PR decay at energies above the band gap is determined by the carrier relaxation. At the uniform alloy band gap energy (process A in Fig. 4), the main contribution to the reflectance change comes from the carrier dynamics in the extended states. The time-resolved PL measurements have shown that most of the photoexcited holes relax to the sub-band edge states and leave AlInN by transfer to the GaN layer in less than 6 ps. Consequently, the electron dynamics controls the longer decay time component of the band gap PR. A probable process defining the electron lifetime in the extended states is diffusion towards dislocations and nonradiative recombination. In agreement with the TRPL results, the fast, subpicosecond decay component should be assigned to the hole capture into the sub-band edge states. The short decay time implies a high surface density of the localized states, so that little time is required for a hole to find a lower potential site via diffusion. This result confirms the previous observation of the scanning near-field optical microscopy that the size of and the spacing between the carrier localization sites are less than 100 nm. The main candidate for these dense sub-band edge states are the nanoscopic In clusters formed by In atom arrangements around nitrogen atoms.

At lower energies, optical transitions involve the sub-band edge states. Increasing PR decay time with decreasing photon energy can be attributed to the increased contribution of the slow carrier dynamics in the deep sub-band edge states. The decay times of 40–60 ps can either be assigned to electrons in the extended or the localized states, or to the localized holes (process B in Fig. 4).

At still lower photon energies, around 3.9–4.1 eV, the transient abruptly changes sign, and the faster component of the reflection recovery time falls to \( \sim 1 \) ps. Such an abrupt change of the PR character has never been observed before in semiconductor heterostructures and deserves a longer discussion.

Since the absorption length at energies below the uniform alloy band gap is larger than the AlInN layer thickness, the PR signal probes AlInN and GaN layers, as well as the interface. The change in the PR sign might occur due to the band gap renormalization (not relevant for AlInN and GaN for the used power densities), the induced absorption, or the electro-optic effect. In the latter case, the PR signal is determined by the change of the interface electric field. For the negative \( \Delta R \) sign, the field should increase after the pump. This may occur due to photoexcited electrons drifting to the interface from the bulk of GaN (and the holes being swept by the field deeper into the structure).
FIG. 5. PR transients at 3.84 eV for 10 nm Al$_{0.86}$In$_{0.14}$N/GaN structure and GaN.

and/or the holes excited in AlInN close to the interface. To distinguish whether the negative $\Delta R$ is determined by the electro-optic effect or by the induced absorption involving localized and/or defect states, the experiment has been repeated on the Al$_{0.86}$In$_{0.14}$N/GaN heterostructure with just a 10 nm thick AlInN layer. For this structure, the contribution from the induced absorption to the PR signal should be nearly an order of magnitude smaller than that for the thicker 95 nm AlInN layer. On the other hand, the field effect should be similar, since the interface for both samples is nearly the same. Fig. 5 shows the PR transient of the 10 nm sample along with the transient of the GaN epilayer. The sharp positive peak in the transient of the AlInN/GaN sample resembles that observed for the GaN layer and can be attributed to the carrier thermalization and cooling in GaN. The following negative signal, of similar amplitude as for the 95 nm AlInN structure, seems to have the same origin as in the thicker AlInN samples, namely, the electro-optic PR from the interface.

The threshold-like change of the PR sign requires a mechanism that is efficient for photon energies below 3.9–4.1 eV, but not above. The electron drift from the GaN layer to the interface should not experience a spectral threshold at energies around 4.0 eV, since these states are well above the GaN band gap. On the other hand, holes, generated at these photon energies in AlInN, can be trapped. If one assumes that the holes in question are generated close to the interface by optical transitions from the sub-band edge valence band states to the extended conduction band states, the holes might remain localized while electrons would rapidly transfer to the well or to the bulk of the AlInN layer (process C, Fig. 4). The energy of such hole states above the edge of the uniform alloy valence band would be 0.3–0.4 eV. However, the hole localization in the deep sub-band edge states of AlInN is weak. Thus, the localized holes would, via tunneling between clusters and/or diffusion via extended conductive defects eventually move into the GaN layer and drift away from the interface. This process would account for the PR transient decay. One could question why this mechanism is relevant for the sub-band edge hole states with energies larger than 0.3–0.4 eV and not smaller (as evidenced by the positive transients measured for the higher photon energies). A possible explanation could be a spatial distribution of the In clusters, as illustrated in Fig. 4. If clustering is stronger close to the interface, the deep valence band states will be located in this region. For carrier generation from the shallower states in the bulk of the AlInN layer, electrons would have a much smaller probability to transfer into the well because of the larger distance to the interface and Coulomb attraction to the photoexcited holes. In such a case, the electrons and holes would not separate in space, the interface field would remain unaffected, and the carriers would affect PR via the state filling and not via modification of the interface field. Indirectly, this is confirmed by the measurements on the 10 nm AlInN structure, for which no positive slow transients, resembling those measured at energies between 4.1 and 4.5 eV for the 95 nm sample, have been observed.
At still lower energies, the transient amplitude increases and the PR signal changes sign again (Figs. 2 and 3). The energy difference between the uniform alloy band gap and the energy of the second sign reversal is about 0.8 eV. At these energies, AlInN absorption may be too small to generate a negative electro-optic PR transient. Instead, PR at these photon energies should be attributed to bleaching of the GaN absorption (process D, Fig. 4). The strongly increased PR amplitude confirms that now the signal is determined by a layer with a large number of states. The double exponential decay with the time constants of 10–20 ps and ~300 ps, respectively, corresponds to the times of the GaN PL decay. The short decay time reflects the photoexcited electron and hole separation by the interface field, whereas the slow process is linked to the nonradiative recombination.

Some PR traces exhibit absorption oscillations with a period of 6.1 ps in the sub-band gap region (see the 4.18 eV curve in Fig. 2). These oscillations occur due to the reflectance perturbation by a coherent acoustic phonon pulse. This pulse is generated by the relaxation of carriers excited by the pump pulse at AlInN/GaN interface and propagating through AlInN. The oscillation period estimated as\(^{23}\) \(T \approx \lambda / (2Cn)\) is 6.6 ps. Here \(\lambda\) is the probe wavelength in vacuum, \(C\) is the sound velocity along the polar axis in Al\(_{0.82}\)In\(_{0.18}\)N (9930 m/s, evaluated as the linear interpolation between the data for AlN and InN) and \(n\) (2.27) is the refractive index. This value is in reasonable agreement with the experiment. For the acoustic pulse propagation in GaN, the period is somewhat longer, 8 ps.\(^{23}\) In our case, however, the main contribution to the oscillations originates from the AlInN layer, because of the strong above-band gap absorption in GaN. Interestingly enough, oscillations with a similar 5.5–6 ps period have been observed in the PR transients of AlGaN.\(^{17}\)

In conclusion, time-resolved photoreflection has been used to study carrier dynamics above and below the uniform AlInN alloy band gap in AlInN/GaN heterostructures. For energies at the band gap, the subpicosecond signal decay indicates an ultrafast carrier relaxation from the extended to the sub-band edge states. Such transfer is only possible if these states are dense and located close to each other. Hence, this finding supports the model attributing the sub-band edge states to the valence band of In-rich nanoscopic clusters. At lower energies, a complicated dependence on the PR amplitude and decay time is caused by filling of the localized states and modulation of the interface field by the photoexcited carriers. Most likely, the hole generation in and escape from the deep sub-band edge states close to the interface govern the reflectance dynamics. Overall, the numerous and dense In cluster-related valence band states seem to dominate the localized state spectrum.

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