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Highly polarized photoluminescence and its dynamics in semipolar $(20\bar{2}\bar{1})$ InGaN/GaN quantum well

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Very high polarization degree of 0.98, considerably larger than theoretical predictions, has been measured in $(20\bar{2}\bar{1})$ $In_{0.24}Ga_{0.76}N$/GaN quantum well by low temperature photoluminescence. With increasing temperature, the polarization degree decreases due to thermal population of the excited valence band level. This effect suggests an accurate method to determine the interlevel energy, which, for the studied well, is 32 meV. Time-resolved photoluminescence measurements set radiative recombination times between 2 and 12 ns for temperatures from 3 to 300 K. Nonradiative recombination was found to be slow, over 2 ns at 300 K, taking place via traps with activation energy of 0.19 eV. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4869459]

Semipolar InGaN/GaN quantum wells (QWs) have recently emerged as an alternative to QWs grown on polar and nonpolar planes for fabrication of light emitting diodes (LEDs) and lasers.[1] Compared to the polar structures, the semipolar ones experience smaller intrinsic electric fields, caused by the difference in spontaneous and piezoelectric polarizations in the barrier and well layers. In the semipolar InGaN QW family, $(20\bar{2}\bar{1})$ QWs stand out because of their low efficiency droop.[1] Advantage of the $(20\bar{2}\bar{1})$ QWs over the nonpolar ones is a larger In uptake under the same growth conditions,[2] which leads to longer emission wavelengths, including those in the green spectral region. Besides, the $(20\bar{2}\bar{1})$ QWs experience weaker carrier localization, which results in narrower widths of the emission spectra,[3] advantageous for laser applications.

Light emission from InGaN QWs grown on nonpolar and semipolar planes is, in general, elliptically polarized. Usually, the polarization properties are defined by the degree of linear polarization, $\rho = (I_\perp - I_\parallel)/(I_\perp + I_\parallel)$, where $I_\perp$ and $I_\parallel$ are light intensities for electric field directions perpendicular to the polar **c** axis and parallel to it or its projection to the growth plane (Fig. 1). For the $(20\bar{2}\bar{1})$ plane, the **c** axis projection has $[\bar{1}01\bar{4}]$ direction and makes a 15° angle with the **c** axis. The **E**⊥**c** direction is parallel to $[\bar{1}2\bar{1}0]$. The polarization degree is an important design parameter for LEDs used in backlighting of liquid crystal displays; it also defines the preferential cavity direction in lasers. The degree of the optical polarization is primarily defined by properties of the band structure,[4] however, it can be significantly affected by carrier localization.[5,6] Thus, data on the polarization degree is also important for studies of basic properties and localization effects in ternary nitrides of nonpolar and semipolar orientations. As yet, theoretical models do not always reproduce the experimental data on interlevel energies and optical polarization features of InGaN QWs.[4] Consequently, detailed experimental information is required for the refinement of models and material parameters. The available data on the polarization degree is often limited to room temperature values. At room temperature, transitions to several valence band levels of different symmetry contribute to the polarization degree of the emitted radiation. Since the interlevel energies and symmetry properties of the transition matrix elements are not always known or can be reliably calculated, low temperature data, depending only on transitions between the ground levels, is desirable.

For efficient emitters of polarized radiation, not only the polarization degree, but also the radiative recombination rate is of importance. A large recombination rate allows for higher injection currents, higher gain, and larger light output. A low radiative recombination rate, on the contrary, makes a device more vulnerable to the nonradiative recombination. The radiative recombination rate in QWs grown on other than nonpolar planes is affected by the intrinsic electric field via reduced electron and hole wave function overlap. In polar InGaN QWs, this leads to long radiative recombination times.[7] Even though semipolar QWs experience smaller fields than the *c*-plane QWs, the field still affects the radiative lifetime, especially in wider QWs. Since there is little data on the recombination dynamics in $(20\bar{2}\bar{1})$ InGaN QWs,[8] this material property should be properly explored before the full potential of $(20\bar{2}\bar{1})$ QWs for LED and laser applications can be assessed. Here, one should note that though apparently identical, $(20\bar{2}\bar{1})$ and $(20\bar{2}1)$ plane QWs have different properties,[9] and data obtained for one kind of QWs cannot be automatically transferred to the other one, especially for light emitting devices.

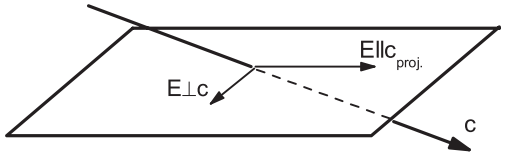

FIG. 1. Schematic diagram of $(20\bar{2}\bar{1})$ plane and optical polarization directions with respect to the **c** axis of the wurtzite structure.

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In this work, the polarization degree and the radiative lifetime have been studied in a high quality $(20\bar{2}\bar{1})$ single InGaN QW structure by time-resolved photoluminescence (PL). Surprisingly, the low temperature PL is nearly 100% linearly polarized, which contradicts the theoretical predictions. The radiative carrier lifetimes were found to be relatively short, confirming the high potential of $(20\bar{2}\bar{1})$ QWs for photonic applications.

The studied structure was grown by metal organic chemical vapor deposition on bulk $(20\bar{2}\bar{1})$ plane GaN substrate, provided by Mitsubishi Chemical Corporation. The dislocation density of the substrate, as specified by the supplier, was of the order of $10^6\,\mathrm{cm}^{-2}$. The structure consisted of a 1 μm undoped GaN template layer, a 3 nm $In_{0.24}Ga_{0.76}N$ single QW and a 10 nm GaN cap layer. The QW growth temperature was 855 °C. QW PL spectra are spatially uniform, as recently evidenced by scanning near-field optical microscopy (SNOM) measurements.[3]

Time-resolved PL measurements were carried out using a frequency doubled femtosecond Ti:sapphire laser (150 fs pulse duration, 400 nm central wavelength, 76 MHz pulse repetition frequency) and a spectrometer – streak camera system. Temporal resolution of the experiments was 10 ps. For 400 nm excitation, carriers were generated only in the QW. The sample temperature was varied between 3 and 300 K with the help of a closed cycle He cryostat. PL polarization was analyzed by placing a linear film polarizer with a high extinction ratio just outside the cryostat window. The average photoexcited power density was between 0.1 and 3 mW, which corresponds to the peak photoexcited carrier density of 1×10^{10} and $3 \times 10^{11}\,\mathrm{cm}^{-2}$, respectively.

In unstrained nitride semiconductors with a wurtzite structure, three uppermost valence bands at the Brillouin zone center have symmetry Γ_9, Γ_7^{upper}, and Γ_7^{lower}, labeled heavy hole (HH), light hole (LH), and split-off (SO) hole bands.[10] The HH and LH bands are a mixture of p_x and p_y atomic orbitals with wave functions of the character $|X \mp iY\rangle$, and the split-off band is formed of p_z orbitals with the $|Z\rangle$-like wave function, where the **z** direction is parallel to the **c** axis. When an InGaN layer is grown on the *c*-plane GaN, the layer experiences an equibiaxial compressive stress, and symmetry and polarization properties of the valence bands are unchanged. If an InGaN layer is grown on a nonpolar or a semipolar plane, the **x** and **y** directions are no longer equivalent, and the original valence band states are strongly modified. For the nonpolar case, the original $|X \mp iY\rangle$ HH and LH states become $|X\rangle$ and $|Y\rangle$-like. The in-plane compression along the **x** axis induces expansion along the **y** axis; as a result, the $|Y\rangle$-like band is shifted in energy below the $|Z\rangle$-like band. Optical transitions between the conduction and the three valence bands become polarized, with the ground state transition polarization perpendicular to the **c** axis. For semipolar planes, the situation is even more complicated due to the shear component of strain.[11] $\mathbf{K}\cdot\mathbf{p}$ models predict that the valence bands in semipolar layers have contributions from all three bands (HH, LH, and SO), which reflects in the selection rules of optical transitions. The top most valence bands in semipolar layers are labeled A and B.[4] Transitions into these bands are polarized predominantly perpendicular and parallel to the projection of the **c** axis to the growth plane. Quantum confinement modifies the valence band states, however, the two upper-most levels are considered to originate from the A and B bands.[4,12]

The polarization degree for transitions between the first conduction and valence band levels in $(20\bar{2}\bar{1})$ InGaN QWs has been calculated in several works. According to Yamaguchi,[13] the polarization degree has a very strong dependence on the QW width and strain; thus, the calculation results are difficult to compare to the experimental values. A more direct comparison of the experimental data can be made with calculations by Schade *et al.*,[4] who have reported a polarization degree of 0.90. In Ref. 14, for 3 nm $In_{0.24}Ga_{0.76}N$/GaN QW, the calculations provided a polarization degree of 0.6.

Figure 2 shows 3 K and 270 K time-integrated PL spectra for polarizations perpendicular to the **c** axis and parallel to its projection in the QW plane. Emission, especially at low temperature, is strongly polarized. At 3 K and low photoexcited carrier density ($\sim 1 \times 10^{10}\,\mathrm{cm}^{-2}$), which are the required conditions to avoid population of the higher QW levels, the polarization degree is as high as 0.98. Moreover, the measured value can be considered as the lower limit of the intrinsic polarization degree, since some PL depolarization might occur due to light scattering from the rough back surface of the sample, even though that surface was painted with black ink.[15] Another effect, lowering the polarization degree, namely, carrier localization, should have a minor influence in our case, since for the studied sample emission primarily occurs from the extended states,[3] and carrier transfer to the localized states in such structures at low temperature is inhibited.[16]

Remarkably, the measured polarization degree is even larger than most of the values reported for nonpolar QWs,[5,6,10] which, according to some calculations,[4] should emit completely linearly polarized light. Similarly high polarization degree as in our $(20\bar{2}\bar{1})$ QW has recently been reported for *m*-plane InGaN QW,[15] however, in that case elaborate corrections for light depolarization due to scattering from the back sample surface had been applied. In our work, the 98% polarization degree was obtained directly from the measurements.

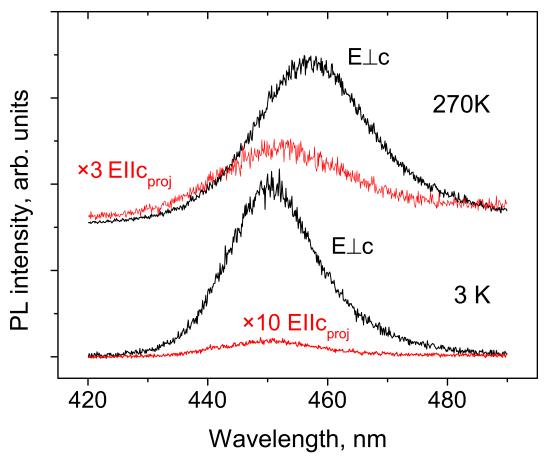

FIG. 2. Time-integrated 3 K and 270 K spectra for $\mathbf{E}\perp\mathbf{c}$ and $\mathbf{E} \parallel \mathbf{c}_{proj}$ polarizations.

The measured polarization degree is larger than predicted by the theoretical calculations.[4] Such occurrence, however, is not unique. Larger than calculated polarization degree has been experimentally observed in semipolar $(11\bar{2}2)$ QW as well.[4] The authors have suggested that this may be because of limitations of the strain model used in the calculations and/or uncertainties in deformation potential values. Our very high value of the polarization degree also calls for revision of the parameters used in $\mathbf{k}\cdot\mathbf{p}$ calculations. In particular, our measurement results seem to indicate that mixing of the $|X\rangle$ and $|Y\rangle$-like valence bands with the $|Z\rangle$-like band in the formation of the upper-most valence band level in the QW is marginal, if any.

With increased temperature, the polarization degree decreases (Fig. 3) due to the thermal population of the second hole level. This effect can be used to determine the interlevel energy between the valence band states. As the second hole level is populated according to the Boltzmann statistics, the temperature dependence of the polarization degree can be expressed as $\rho = \rho_0\left(1 - A\exp\left(-\Delta E/(kT)\right)\right)$, where ρ_0 is the degree of polarization at 0 K and A is the coefficient depending on the matrix elements for transitions to the first and second hole levels. For equal matrix elements, $A = 0.5$. The fit of the experimental data to the given equation (Fig. 3) provides the interlevel energy of 32 meV. It coincides with the calculated value of 32 meV for 3 nm $(20\bar{2}\bar{1})$ $In_{0.24}Ga_{0.76}N$ QW[9] and is very close to the energy difference of 30 meV estimated from the high temperature PL peak positions for different polarizations (Fig. 2). The method to determine the interlevel distance from the second hole level population might be superior to the way of estimation from the high temperature PL spectra. Firstly, this is because of the large inhomogeneous broadening, typical for ternary nitride QWs, and secondly, because of difficulties determining the PL peak positions related to the different transitions in QWs with a small polarization degree.

Degree of optical polarization is a measure not only of transition matrix elements and interlevel energies. Its dynamics may provide information on the carrier transfer from extended to localized states, and on symmetry properties of the localized states.[6] Therefore, dynamics of the polarization degree was also investigated for the studied $(20\bar{2}\bar{1})$ QW. The temporal dependence of the polarization degree was calculated from PL transients measured for $\mathbf{E}\perp\mathbf{c}$ and $\mathbf{E} \parallel \mathbf{c}_{proj}$ polarizations. Contrary to previous measurements on *m*-plane QWs,[6] the polarization degree did not show any temporal change, neither at low nor at high temperatures. This shows that the localized states in the studied QW have the same symmetry properties as the extended states. This allows attributing the origin of the localized states to small alloy composition fluctuations rather than to deep quantum dot like states, which, due to the different symmetry, would have different polarization properties.[5] This observation is consistent with the SNOM data showing large areas of uniform potentials and a small localization depth.[3]

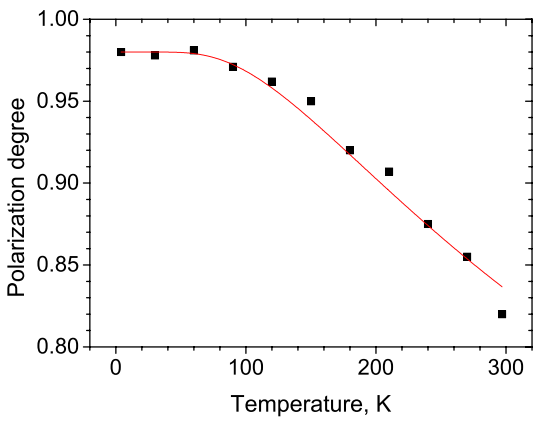

FIG. 3. Temperature dependence of the polarization degree.

Decay of the PL intensity (inset to Fig. 4) has a biexponential shape with the short decay time of about 150 ps and the longer one – in the ns range. The ratio of weight coefficients in the double exponential fit for the long and short decays is between 3 and 4, thus, the longer decay is prevailing. The shorter decay time reflects de-screening of the built-in electric field.[17] The slower PL decay with time τ_{PL} is determined by recombination; it depends on the radiative τ_r and nonradiative recombination τ_{nr} times according to $1/\tau_{PL} = 1/\tau_r + 1/\tau_{\mathrm{nr}}$. In the following, we will focus on this longer PL decay time. Temperature dependencies of the long decay time and the transient amplitude (Fig. 4) allow evaluation of the radiative and the nonradiative recombination times.[18,19] This evaluation is based on an assumption that at low temperatures carrier trapping to deep centers during the first few tens of ps is negligible. For the high quality and spatially uniform QW grown on a native GaN substrate with a low dislocation density, such an assumption is justified.

Figure 5 shows the temperature dependence of the radiative and the nonradiative recombination times. The radiative lifetime changes from 2 ns at low temperature to 12 ns at room temperature. The nonradiative recombination prevails at temperatures larger than 200 K. Still, the nonradiative lifetime at all temperatures is long, over 2.4 ns. At low temperatures (<100 K), the temperature dependence of the radiative recombination time is weak, and the time is determined by the lifetime of localized excitons.[20] In the middle temperature range (100–240 K), recombination is dominated by free excitons. With temperature, the thermal distribution of excitons increases their in-plane *k* values including those outside

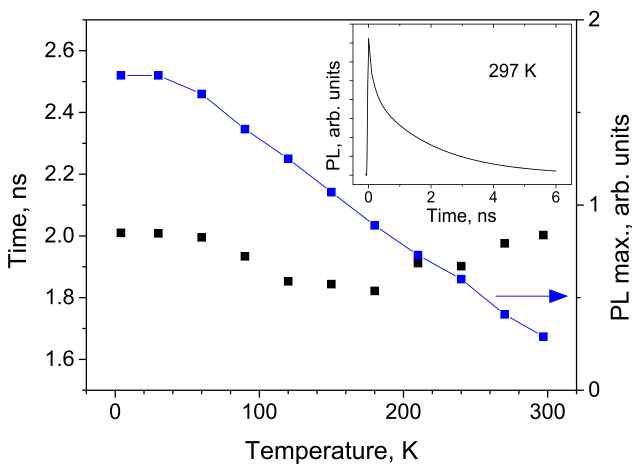

FIG. 4. Temperature dependence of the longer PL decay time and the transient amplitude (right axis). The inset shows 297 K PL transient for $\mathbf{E}\perp\mathbf{c}$ polarization (linear scale).

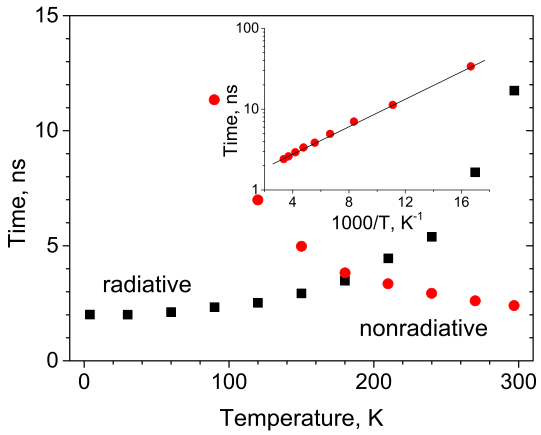

FIG. 5. Temperature dependence of the radiative and nonradiative recombination times. The inset shows Arrhenius plot for the nonradiative recombination time.

the light cone, and the radiative recombination time experiences a linear rise.[20] At high temperatures, increased contribution of free carriers and thermal spread of heavy holes to wave vectors exceeding those of electrons further enhances the time of the radiative recombination.

The radiative lifetime can be compared to values for QWs of other crystallographic orientations. In nonpolar *m*-plane QWs, the radiative lifetime is short, about 0.5 ns at low temperature.[19] In *m*-plane QWs, there is no intrinsic electric field to separate electrons and holes, and prolongation of the radiative lifetime may occur only due to carrier localization. In polar *c*-plane QWs, the intrinsic electric field is strong, >2 MV/cm.[7] For QWs of similar thickness and composition as our semipolar QW, the low temperature radiative lifetime is about 20 ns.[7] For $(20\bar{2}\bar{1})$ QWs the field is weaker, of the order of 0.1 MV/cm.[1,7] Nevertheless, its influence on the radiative lifetime, compared to the nonpolar case, is still substantial. On the other hand, due to the spatial uniformity of the alloy composition in the studied sample and the small localization potential depth,[3] the radiative recombination times evaluated in our study should be little affected by the localization and should be close to intrinsic values.

The temperature dependence of the nonradiative recombination time allows estimating the main channel of the nonradiative process. An Arrhenius-type plot (inset to Fig. 5) shows a single-exponential dependence of the nonradiative recombination time on the inverse temperature, which provides an activation energy of 0.19 eV. This value is much larger than a few tens of meV, typically observed in polar[20] or nonpolar InGaN QWs.[19] In the mentioned studies, the activation process was assigned to thermal exciton emission out of the localization sites and overcoming potential barriers surrounding deep recombination centers. In the case of $(20\bar{2}\bar{1})$ QW, the activation energy is too large for such processes, however, it is close to the reported values of carrier traps involving N vacancies[21] and C acceptors.[22] The absence of the shallow, localization related activation process is consistent with the observations of spatially uniform band potentials and weak localization in $(20\bar{2}\bar{1})$ QWs.[3]

In conclusion, time- and polarization-resolved PL measurements were performed on 3 nm wide $In_{0.24}Ga_{0.76}N$/GaN QW in a wide temperature range. A very high linear polarization degree of 0.98, considerably exceeding the theoretical predictions, was observed. The temperature dependence of the polarization degree allowed determining the interlevel energy for the QW valence band levels. The radiative recombination times, which are in the interval from 2 to 12 ns for temperatures between 3 and 300 K, were estimated from PL transient amplitudes and decay times. The nonradiative recombination was found to be slow, taking place via traps with the activation energy of 0.19 eV.

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