S shape in polar GaInN/GaN quantum wells: Piezoelectric-field-induced blue shift driven by onset of nonradiative recombination

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(Received 16 June 2014; revised manuscript received 15 October 2014; published 14 November 2014)

In this paper, we critically review the usual explanation of the blue shift in the temperature dependence of the light emission, which is commonly observed for polar GaInN/GaN quantum wells at intermediate temperatures. We demonstrate that this blue shift is not necessarily caused by a thermally induced change of occupation in an inhomogeneously broadened density of states. Instead, different energy dependencies of radiative and nonradiative lifetimes may lead to an energy dependence of the internal quantum efficiency strongly influencing the peak position of the luminescence: the piezoelectric fields within the quantum well induce an approximately exponential relationship between the electron-hole overlap matrix element for the radiative transition between the lowest quantized states and the respective transition energy. Via time-resolved photoluminescence spectroscopy, we observe a corresponding exponential energy dependence of radiative lifetimes at low temperatures and almost energy independent nonradiative lifetimes toward room temperature throughout the emission of polar single quantum wells. An analytical model is demonstrated, predicting a significant blue shift of several 10 meV for polar and a negligible shift for nonpolar GaInN/GaN quantum wells at the transition from high to low internal quantum efficiency due to the energy-dependent competition between radiative and nonradiative recombination processes. This model consistently explains the lack of a blue shift in nonpolar quantum wells grown on *m*-plane bulk GaN as well as a reduced characteristic temperature for the onset of the blue shift after artificial defect generation via argon ion implantation.

DOI: 10.1103/PhysRevB.90.205302 PACS number(s): 78.66.Fd, 78.67.De, 72.20.Jv

I. INTRODUCTION

GaInN quantum wells embody an enormous field of research due to the high potential for application in optoelectronic devices. Although this field has been widely investigated over the recent decades, several fundamental questions are not fully understood. One open question is related to the importance of carrier localization being caused by potential fluctuations within the quantum well plane due to fluctuations of the atomic composition and the well width. This has a significant impact on the optical properties [1–6]. As a consequence, the concepts of carrier localization in a disordered system and transport between localization sites [7,8] are often applied to GaInN quantum wells [9–12]. Specifically, the potential fluctuations are evident by a strong inhomogeneous broadening of several 10 meV to more than 100 meV, which is typically observed in luminescence spectra of GaInN/GaN quantum wells [13,14]. Additionally, an "S"-like shaped red-/blue-/redshift in the temperature dependence of the peak emission energy is linked to potential fluctuations and carrier localization as well [15,16].

According to the usual explanation, the blue shift in the intermediate part of this S-shape is caused by a Boltzmann-like occupation of states, leading to an increased population of higher energy states (within an ensemble of states) with increasing temperature [15,17,18]. An evaluation of this blue shift is frequently used to determine a characteristic thermal activation energy for a lateral carrier transport, as the higher energy states are interpreted as "mobile" [15,19–21]. A fundamental problem arises, as a Boltzmann-distributed occupation of localized states according to the lattice temperature can only be established, if spatial transport processes of carriers between localized states are fast compared to the carrier lifetimes. This means that carrier diffusion lengths need to be large, which is in contradiction to the assumption of localized carriers. Via numerical carrier hopping models, it has been shown that the competition between carrier transport and recombination processes may explain both the low-temperature redshift as well as the blue shift at intermediate temperatures [22]. However, a detailed understanding of experimental data using hopping models is rather difficult, as reasonable values for model parameters like the attempts-to-hop frequency—a crucial parameter when comparing the hopping and recombination probabilities—are hard to predict *a priori*.

Detailed investigations on the temperature dependence of GaInN/GaN quantum well photoluminescence tend to show that the thermally induced blue shift is accompanied by a drop of emission intensity within the same temperature regime [10,16,23]. Common explanations of this coincidence claim that both the blue shift and the onset of nonradiative recombination are symptoms of a thermally activated carrier redistribution from low-energy "localized" states (carriers are unable to reach the nonradiative centers) to high energy "mobile" states (carriers are able to reach the nonradiative centers) [10,24].

In this paper, we demonstrate that both the blue shift and the concomitant drop of intensity in polar GaInN quantum wells can be understood within a completely different picture: we propose an energy-dependent competition between radiative and nonradiative recombination processes being

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driven by an energy dependence of radiative lifetimes caused by piezoelectric fields. An onset of energy independent nonradiative recombination processes may shift the emission spectra toward higher energies at intermediate temperatures. Thus the commonly observed thermally induced blue shift in polar GaInN/GaN quantum wells can be understood even without regard to a thermally induced redistribution of carriers. Such a mechanism is neglected in the discussion of the S shape up to now. Furthermore, we demonstrate key experiments in order to identify the dominant contribution to the blue shift.

II. EXPERIMENTAL DETAILS

We investigate the photoluminescence properties of *c*- and *m*-plane GaInN/GaN quantum well structures grown via metal-organic vapor phase epitaxy (Aixtron AIX200RF). The *c*-plane samples were grown on sapphire substrates, consisting of a 2-μm silicon-doped GaN buffer, an optically active region and an unintentionally doped GaN capping layer. The optically active region consists of unintentionally doped GaInN quantum wells and GaN barriers, either as single quantum well (SQW) or fivefold multiple quantum well structures (MQW). Threading dislocations are intentionally decorated by V pits in order to reduce the nonradiative recombination probability [25]. Two nonpolar fivefold GaInN/GaN MQW with different indium content were grown on m-plane bulk GaN substrates with low basal plane stacking fault density. The width of the quantum wells are kept below 3 nm for all samples under investigation.

Argon ion implantation has been performed at room temperature using a Varian Extrion 200 DF4 (150-keV acceleration voltage, implantation current density <100 nA/cm^2, incident angle: 7° with respect to the surface normal to avoid channeling of ions). A nominal thickness of the GaN cap layer of 50 nm has been chosen to match the depth of the quantum well beneath the sample surface with the depth of maximum irradiation induced point defect densities [26]. The implantation doses are kept far below the amorphization threshold by at least three orders of magnitude [27].

The quantum well luminescence was measured via temperature dependent time-resolved photoluminescence spectroscopy using time-correlated single photon counting (PicoQuant PicoHarp 300). A helium bath cryostat and a temperature controlled sample holder was used to vary the sample temperature between 5 and 325 K. The quantum wells were selectively excited (380-nm wavelength, 5-ps pulse length, 5–10 nJ/cm^2 incident energy density per pulse, 4-MHz repetition rate) by the second harmonic of a mode-locked and cavity-dumped dye laser beam (dye: Radiant Dyes Pyridine 2). The dye is optically pumped by the second harmonic of a synchronously mode-locked Nd:YAG laser beam (Spectra-Physics, Model 3800). Using a Jobin Yvon Spex 1680 subtractive double-grating monochromator and a microchannel plate photomultiplier (Hamamatsu R3809U-02, 25-ps time resolution), the emitted light is detected.

Decay time constants have been determined by an exponential fit of the initial part of the intensity decay, in which the intensity drops from maximum intensity to $1/\sqrt{e}$ (approximately 60%) of the maximum intensity, as shown in Fig. 1. On the one hand, this allows a comparable handling

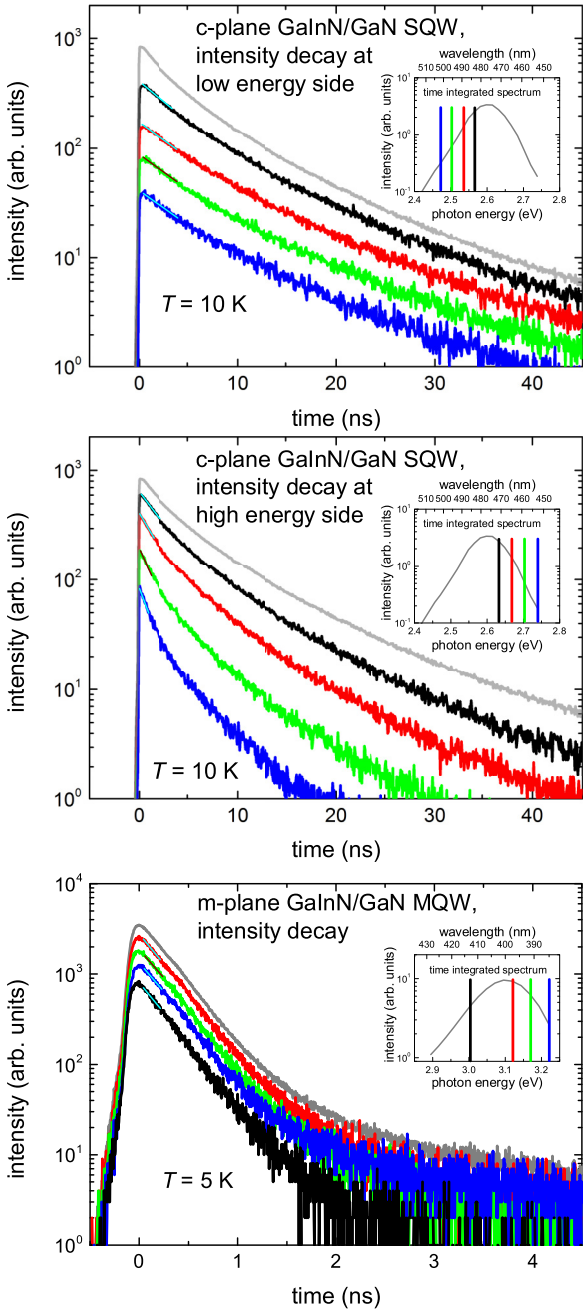

FIG. 1. (Color online) Intensity decay after pulsed excitation at different luminescence detection wavelengths for a *c*-plane SQW (InN mole fraction: nominally 20%, well width: nominally 2.3 nm) and a *m*-plane MQW (InN mole fraction: ≈14%, well width: ≈1.6 nm). Exponential fits of the initial part of the decay are shown in cyan and brown. The decay of the total quantum well emission is shown in gray (vertically shifted for clarity). The insets show the time integrated luminescence at 10 K. Vertical lines indicate the spectral positions of the intensity decays as a definition of the color code.

of the nonexponential behavior, which is commonly observed in GaInN quantum wells. As the laser pulse length is much shorter than carrier lifetimes, the initial excess carrier densities is almost temperature-independent. Thus the time constants taken from the initial decay are related to comparable values of the total excess carrier densities on the other hand.

Peak emission energies have been determined by fitting a Gaussian function that is accompanied by three LO phonon replicas at the low-energy side (LO phonon energy assumed as 92 meV [28]) to the photoluminescence spectra. Structural properties of MQWs were investigated via high-resolution x-ray diffractometry, allowing an estimation of the quantum well widths L_z and InN mole fraction x_{In} [29,30].

For the growth of nonpolar samples, we preferred MQW rather than SQW structures, as this allows for a structural characterization of the quantum wells via x-ray diffraction. However, in the polar case, the usage of MQW rather than SQW is not beneficial for the optical characterization: due to the piezoeletric fields within the quantum wells, the ground states of electrons and holes are separately localized to the outermost wells of MQW structures. While electron-hole pairs are generated in each quantum well via selective optical pumping, recombination processes within the wells will likely compete with redistribution processes of carriers between the quantum wells.

III. TEMPERATURE DEPENDENCE OF PEAK EMISSION ENERGIES IN POLAR AND NONPOLAR QUANTUM WELLS

Figure 2 illustrates the S-shaped temperature dependence of the peak emission energy typically observed for polar GaInN/GaN quantum well structures [16]. While the redshift at higher temperatures is likely attributed to the temperature dependence of the fundamental band gap [31,32], the behavior at lower temperatures is commonly explained as a consequence of inhomogeneously broadened densities of states. The low-temperature redshift can be understood in terms of a redistribution of carriers being driven by a thermally activated carrier transport toward states of lower potential [17]. The subsequent blue shift at intermediate temperatures is often related to a Boltzmann occupation of localized states [15,17], assuming

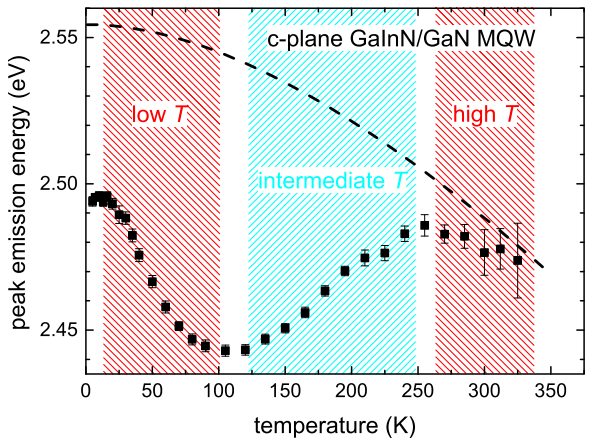

FIG. 2. (Color online) Peak emission energy of a c-plane GaInN/GaN multiple quantum well structure (approximately 20% InN mole fraction and 2.3 nm well width) at different sample temperatures after pulsed excitation, taken from time-integrated spectra: a typical S-shaped behavior is observed. Dashed line serves as a guide to the eyes indicating the temperature dependent shift of the fundamental band gap (quantum confined Stark shift included).

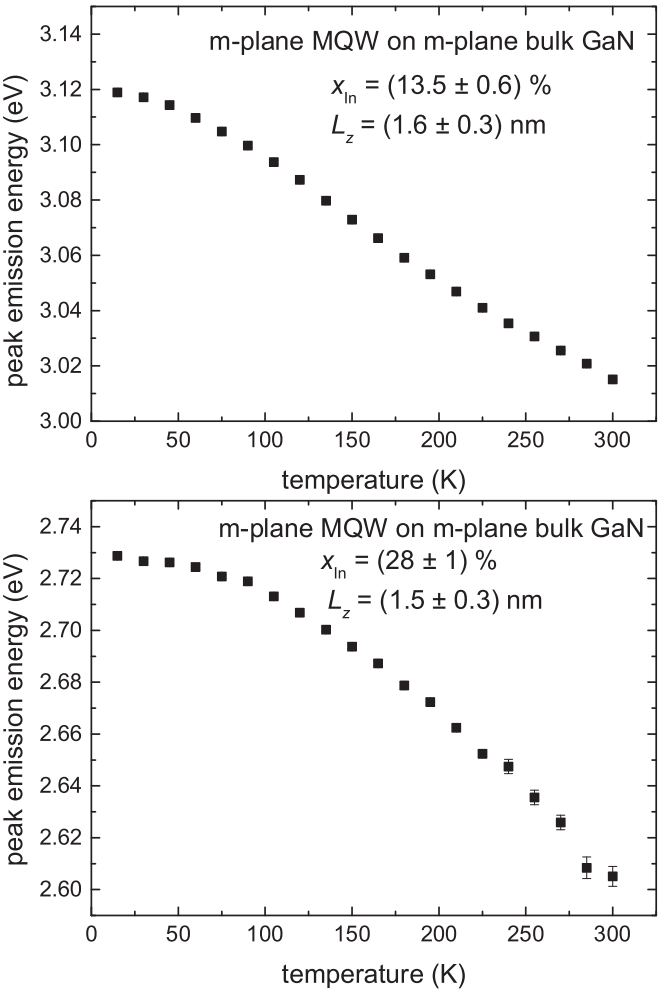

FIG. 3. Temperature dependence of the peak emission energy of m-plane GaInN/GaN multiple quantum well structures grown on m-plane GaN substrates. No blue shift with increasing temperatures is observed.

a rapid redistribution of carriers between laterally separated localized states of different transition energies. This common explanation implies that an S-shaped temperature dependence of peak positions is expected, whenever the density of states is subject to an inhomogeneous broadening.

Keeping this in mind, it is rather surprising that we observe a fundamentally different temperature dependence of the peak emission for nonpolar quantum wells as demonstrated in Fig. 3. Although there is a significant inhomogeneous broadening (low temperature full width at half maximum of 170 meV for the less indium containing sample and 190 meV for the other), the peak emission energy monotonically decreases up to room temperature. For the less indium containing sample, the temperature dependence of the peak emission energy is slightly convex above 150 K. This indicates that the redshift cannot be solely attributed to the temperature dependence of the bulk band gap, which is expected to be a concave function [31,32]. It is possible that the convex behavior is a consequence of a thermal redistribution of carriers from higher to lower energy states—similar to the low temperature redshift in polar wells—which may give an additional contribution

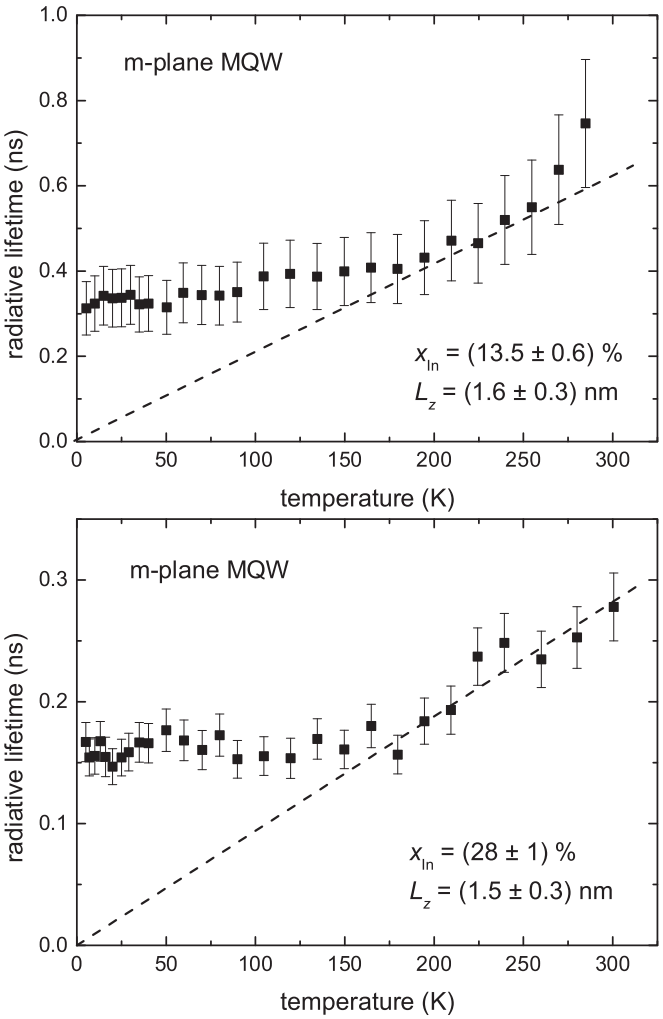

FIG. 4. Temperature dependence of radiative lifetimes of *m*-plane GaInN/GaN multiple quantum well structures grown on *m*-plane GaN substrates. A linear increase above 150 K indicates a two-dimensional free-carrier behavior.

to the observed redshift. When this additional contribution is reduced at higher temperatures (e.g., due to a completed thermal redistribution from higher to lower energy states), a convex behavior would not be unexpected. Yet, compared to polar quantum wells, a similar thermally induced blue shift of the emission is not found up to room temperature for nonpolar quantum wells.

One might argue that carriers may be strongly localized even at room temperature preventing an exchange of carriers between localized states [33]. However, the radiative lifetimes determined via time-resolved photoluminescence spectroscopy (see Fig. 4, method explained in Ref. [34]) does not support this argument. A linear increase of radiative lifetimes as a function of temperature above 150 K indicates that two-dimensional *free* carriers dominate the radiative recombination at room temperature here [35,36]. For the less indium containing wells an onset of a superlinear increase occurs which is likely related to a beginning thermal dissociation of free excitons [37]. Obviously, the lack of a blue shift is not caused by localization of carriers up to room temperature. This significant difference compared to polar quantum wells suggests that the S shape is predominantly affected by the internal polarization fields rather than a result of Boltzmann statistics.

IV. ENERGY DEPENDENCE OF CARRIER LIFETIMES AND LUMINESCENCE DECAY TIMES

It is well known that pseudomorphically grown polar GaInN/GaN quantum wells are subject to strong piezoelectric fields, causing a redshift of the emission line compared to the field-free well according to the quantum confined Stark effect (QCSE) [38]. This goes along with a severe reduction of the matrix element of the radiative transition, as the electron and hole wave functions are separated toward the opposite sides of the quantum well [39,40]. As structural parameters (quantum well width L_z and indium nitride mole fraction x_{In}) are subject to fluctuations, electronic parameters like the band-gap energy, the energy position of quantum confined states and the overlap matrix element will exhibit lateral fluctuations as well. This ends up in a spatially correlated fluctuation of the transition energy and the radiative transition probability [41]: with increasing L_z, the transition energy is reduced almost linearly on the one hand, whereas the overlap matrix element is reduced slightly stronger than exponentially on the other hand, both as a consequence of the piezoelectric field [39]. The influence of an increase of x_{In} on the transition energy and the overlap matrix element due to changes in the band gap and piezoelectric fields is similar to the influence of L_z, even though the impact on the overlap matrix element is much weaker [41]. Consequently, as different positions within the quantum well plane may differ in x_{In} and L_z, the inhomogeneously broadened quantum well emission spectrum is expected to be governed by an almost exponential energy dependence of the radiative lifetime.

Indeed, a corresponding exponential energy dependence of luminescence decay times after pulsed excitation is observed for polar quantum wells at low temperatures, as shown in Fig. 5. Such an energy dependence is neither expected nor observed for nonpolar (*m*-plane) quantum well structures; as the field component perpendicular to the quantum well plane vanishes, the overlap matrix element is hardly affected by fluctuations of L_z or x_{In}. Instead, the overlap remains high giving rise to short radiative lifetimes compared to the polar case. As thermally activated nonradiative processes are unlikely at low temperatures, this may explain the lower absolute values of the time constants shown in Fig. 5.

It should be mentioned that an exponential energy dependence of decay times is commonly attributed to a redistribution of carriers from higher to lower energy states instead, neglecting an energy dependence of carrier lifetimes [42]. Following this argument, the slope of the exponential asymptote only depends on the inhomogeneous broadening of the electron and hole density of states, whereas the carrier lifetime acts as a scaling factor [42]. Thus it cannot explain the observed fundamental difference in the slopes shown in Fig. 5, as the inhomogeneous broadening of the spectra is not very different.

However, a thermally activated contribution of a redistribution from higher to lower energies is evident from temperature dependent time-resolved photoluminescence. For a polar GaInN/GaN quantum well, the energy dependence of

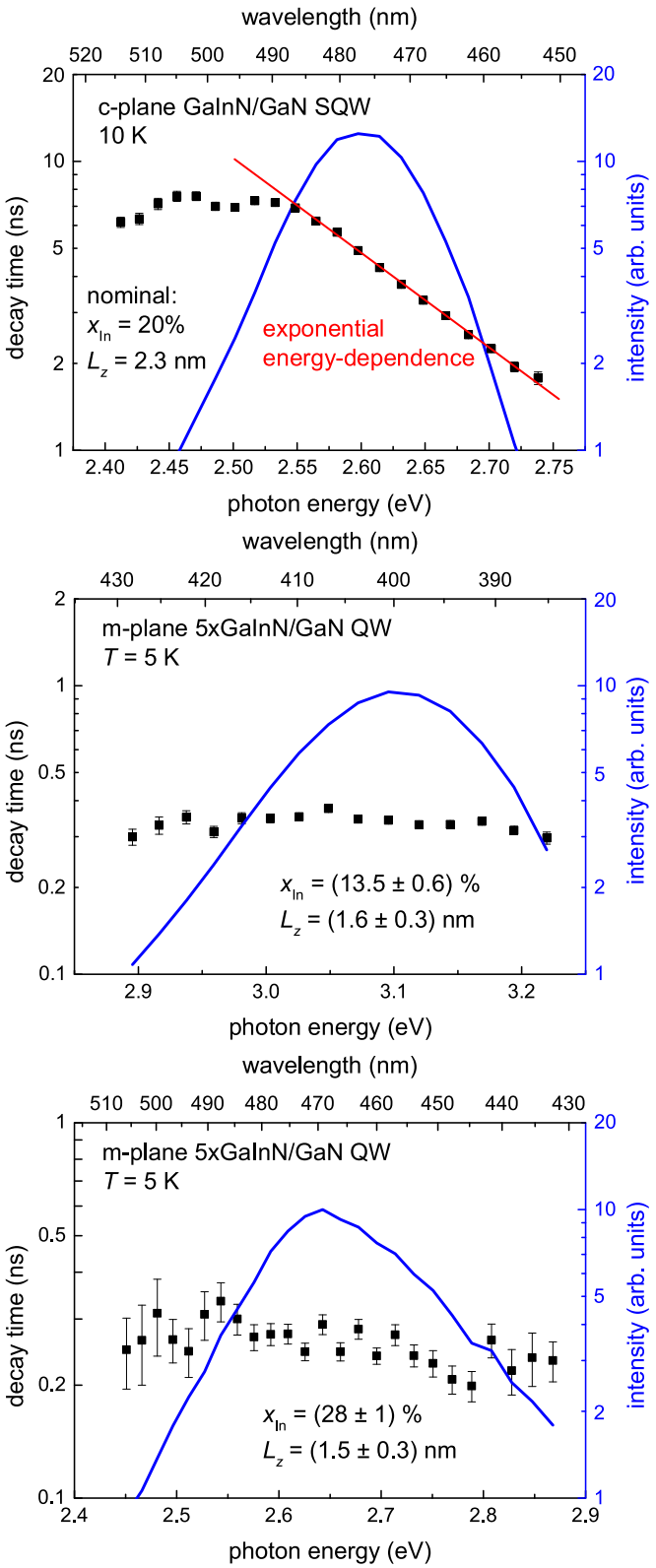

FIG. 5. (Color online) Low-temperature luminescence decay times measured at different spectral positions for a polar and a nonpolar (m-plane) quantum well structure. While the polar structure exhibits an almost exponential energy dependence, decay times are almost constant throughout the whole quantum well emission spectrum in the nonpolar case. The low-temperature time-integrated photoluminescence spectra are shown in blue.

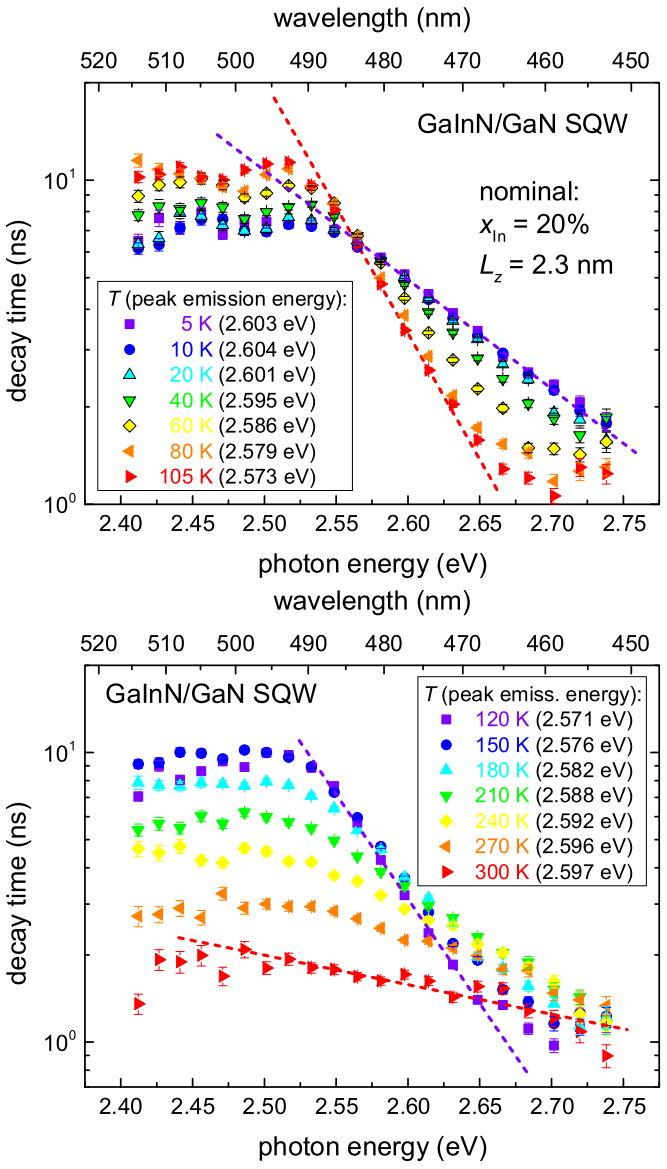

FIG. 6. (Color online) Energy dependence of luminescence decay times of a polar single quantum well structure at different sample temperatures.

decay times at different sample temperatures is shown in Fig. 6. Up to 105 K, the steepness of the slope in a logarithmic plot increases; the shortening of decay times at higher energies and their increase at lower energies can be well understood by a thermal redistribution of carriers toward deeper potentials, accompanied by a redshift of the time integrated emission [17]. This is consistent with recent reports on other polar GaInN/GaN quantum wells [24]. Toward higher temperatures, the steepness of the decay times is quenched, going along with a blue shift of the emission and an over-all reduction of decay times. Such a reduction is consistent with a thermal activation of nonradiative recombination processes [43–47], reducing the effective carrier lifetimes. This observation leads to a crucial question, whether the reduced steepness is caused by either energy independent nonradiative recombination times or sufficiently quick thermal occupation of higher potential states.

V. BLUE SHIFT DUE TO ENERGY DEPENDENCE OF CARRIER LIFETIMES

The total temperature dependence of the decay time spectra illustrates that the recombination is subject to a rather complicated interplay between an energy dependence of recombination times and thermally activated transport mechanisms. This interplay likely governs the emission properties and their temperature dependence. The common explanations of the S shape merely consider the impact of a redistribution of carriers and Boltzmann-occupation of states, neglecting the contribution of energy dependent carrier lifetimes. Thus the underlying assumptions represent an extreme scenario. In the following, the impact of the opposite scenario on the temperature dependence of the emission is discussed; we will neglect the contribution of carrier redistribution and Boltzmann-occupation for the moment, focusing on different energy dependencies of radiative and nonradiative lifetimes. Afterwards, it will be discussed, which one of these extreme scenarios is likely more appropriate to explain the blue shift within the S shape in polar GaInN/GaN quantum wells.

A. Analytical model

Due to the high exciton binding energy in III-nitride quantum wells, we may assume that radiative recombination of bound electron-hole-pairs (excitons) dominates the luminescence—especially at those low temperatures, where the blue shift within the S shape typically occurs [48,49]. In a two-dimensional system, their density of states can be expressed via steplike functions, similar to the single particle density of states. As only excitons with negligible center of mass wave vector can recombine radiatively (momentum of a photon $\ll$ width of Brillouin zone), the linewidth of the luminescence is expected to be sharp. Those states that can contribute to the luminescence thus can be described by a sharp ("δ-function-like") density of states $D(E)$ at an energy E_0, which is given by the energy difference between the deepest confined free electron quantum well state and the deepest confined free hole quantum well state minus the exciton binding energy. The contribution of higher quantized states (both quantum well states and exciton states) shall be neglected in the following.

Let us assume Gaussian fluctuations or variations of the indium composition and the quantum well width around their average values. This gives rise to an inhomogeneous broadening of the density of states given by a convolution of the nonbroadened density of states with a Gaussian function. The total density of exciton states will then be error-function-like. The aforementioned δ-function-like states that can contribute to luminescence changes into a Gaussian function

$$D(E) \propto \exp\left[-\frac{(E-E_0)^2}{2\sigma^2}\right]$$

with a broadening parameter σ.

Furthermore, let us assume that the generation of excess carriers is *evenly* distributed within this density of states: the states cannot be distinguished by their respective carrier generation probability and the carrier generation is temperature-independent. This assumption is in stark contrast to the assumption of a Boltzmann-distribution of carriers, where higher potential states are less likely occupied compared to lower potential states and a significant temperature dependence is included.

The intraband relaxation times shall be fast compared to the carrier lifetimes, leading to an almost instant occupation of states close to the Γ point following an excess carrier generation at higher k values (energy of absorbed laser photon is larger than the energy of the emitted photon). As the intraband relaxation times are in the subpicosecond range [50,51], this assumption is fairly reasonable. Thus one may assume that the generation rate $G(E)$ has the same spectral shape like $D(E)$.

As a key characteristic of this extreme scenario, we neglect a redistribution of carriers within $D(E)$. Furthermore, changes in $D(E)$ (e.g., due to screening of piezoelectric fields) are not considered according to low excitation conditions in the experiments.

The aforementioned exponential energy dependence of the radiative lifetimes can be written as

$$\tau_r(E) = \tau_0 \exp[-s_r(E-E_0)],$$

where τ_0 is the radiative lifetime for $E = E_0$. s_r is a parameter describing the steepness of the slope in a logarithmic plot. Let us furthermore assume that the nonradiative lifetimes

$$\tau_{nr}(E) = \tau_{nr}$$

are energy independent.

The different energy dependence of radiative and nonradiative lifetimes gives rise to a varying internal quantum efficiency $\eta(E)$ throughout the emission spectrum, given by the fraction of the radiative R_r and total rates $R_r + R_{nr}$:

$$\begin{aligned}\eta(E) &= \frac{R_r(E)}{R_{nr}(E)+R_r(E)} \\ &= \frac{\tau_{nr}(E)}{\tau_{nr}(E)+\tau_r(E)} \\ &= \frac{1}{1+\frac{\tau_0}{\tau_{nr}}\exp[-s_r(E-E_0)]}.\end{aligned}$$

This suggests that an onset of nonradiative recombination severely affects the low energy part of the spectrum, where the radiative lifetimes are longest. Here, the nonradiative recombination may easily dominate over the radiative processes, whereas the high-energy contributions still remain highly efficient.

$\eta(E)$ is plotted in Fig. 7, demonstrating the transition between the high efficiency limit with $\eta(E) = 1$ and the low-efficiency limit characterized by an exponential energy dependence of η. In the high-efficiency limit, the peak emission energy coincides with the maximum position of $G(E)$ (i.e., E_0) as a consequence of the equilibrium between the carrier generation rate $G(E)$ and the total recombination rate $R(E) = R_r(E)$.

For continuous wave excitation, the emitted spectrum $I(E)$ is given by $I(E) \propto R_r(E) = \eta(E)G(E)$. An onset of an additional nonradiative recombination channel will displace the emitted spectrum with respect to $G(E)$ along the energy axis. The maximum displacement is reached in the low

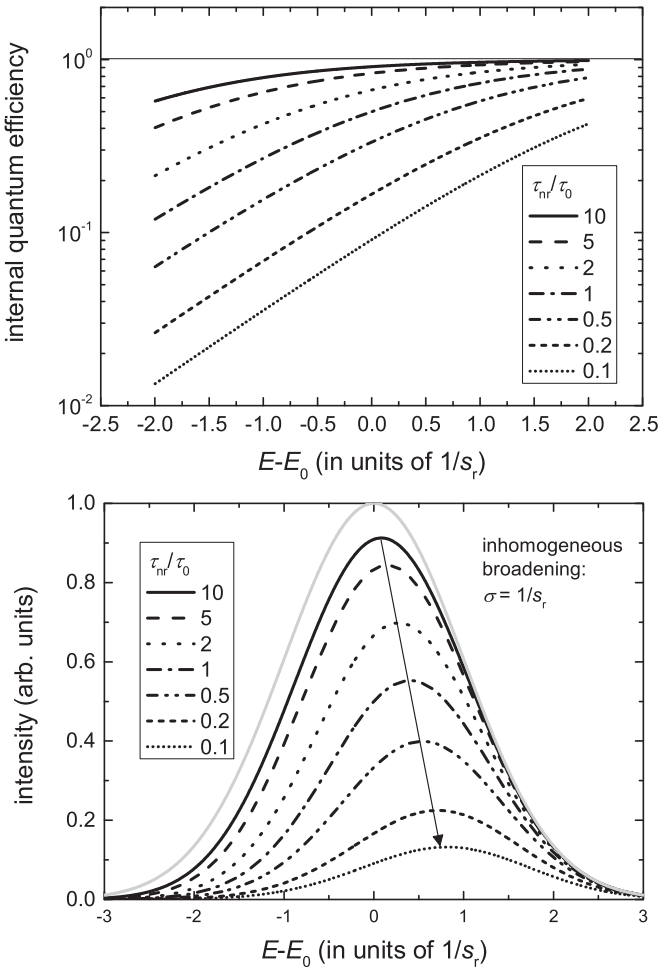

FIG. 7. Energy dependence of internal quantum efficiencies (top) and luminescence intensities (bottom) according to an exponential energy dependence of radiative lifetimes and energy independent nonradiative lifetimes for different values of τ_{nr}/τ_0. The gray line indicates the emission spectrum for purely radiative recombination.

efficiency limit; here, the intensity spectrum can be described via

$$I(E) \propto \exp[s_r(E-E_0)] \exp\left[-\frac{(E-E_0)^2}{2\sigma^2}\right]$$
$$= \exp\left(\frac{1}{2}s_r^2\sigma^2\right) \exp\left\{-\frac{[E-(E_0+s_r\sigma^2)]^2}{2\sigma^2}\right\}$$

using a completion of the square. Thus a blue shift of the spectrum by a total amount of

$$\Delta E = s_r\sigma^2$$

is expected in the low efficiency limit compared to the high efficiency limit. Hence ΔE depends on the energy dependence of the radiative lifetimes and the inhomogeneous broadening of the density of states. This blue shift subsequently occurs at the transition between these two limits, as depicted by the arrow in Fig. 7.

A quantitative estimation of ΔE requires a determination of s_r and σ. Low-temperature photoluminescence spectroscopy at low excitation allows an experimental estimation, as carrier transport processes are likely not thermally activated: σ influences the width of the spectra, whereas s_r contributes to the steepness of the decay time spectra (see, e.g., Fig. 5). As an example, the c-plane single quantum well shown in Figs. 5 and 6 exhibits $s_r \approx 8$ eV^{-1} and $\sigma \approx 50$ meV at low temperatures. Thus a corresponding blue shift by an amount of $\Delta E \approx 20$ meV would be expected. The time-integrated spectra exhibit a blue shift of 26 meV between 120 and 300 K (see Fig. 6)—only slightly larger than the model prediction. The deviations would be somewhat higher, if the thermally induced band gap shrinkage is taken into account. In case of nonpolar structures, the lack of polarization fields perpendicular to the quantum well strongly reduces the variations of the overlap matrix element, leading to negligible values of s_r. Consequently, negligible values of ΔE are expected for nonpolar wells in spite of a significant inhomogeneous broadening, being consistent with the observation in Fig. 3.

It should be noted that in case of continuous wave excitation, this blue shift of the quantum well emission line *is* accompanied by a shift in the occupation of the density of states even in this proposed model. However, this shift is *not* related to a thermally activated carrier transport or Boltzmann-statistics. Instead, it is a consequence of the thermally induced change of the energy dependence of carrier lifetimes $\tau(E,T)$: assuming a dynamic equilibrium between generation and recombination within any energy interval, the density of carriers with an energy between E and $E+dE$ at a temperature T can be described via

$$n(E,T)dE = \tau(E,T)G(E)dE$$
$$= \frac{\tau_r(E,T)\tau_{nr}(E,T)}{\tau_r(E,T)+\tau_{nr}(E,T)}G(E)dE$$

then. This may induce a temperature dependent spectral displacement of the maximum positions of $n(E,T)$ and $G(E)$ relative to each other.

For pulsed excitation (laser pulse shorter than carrier lifetime), a similar behavior is expected for the emission spectra determined via an integration of the intensity decay over the time; here, the spectral distribution of the initially generated excess carrier density $\delta n(E)$ is given by $G(E)$ during the excitation pulse multiplied by the pulse length—independent from the carrier lifetimes. The energy dependence of the time-integrated intensity is then given by $\eta(E) \times \delta n(E)$, leading to the same result as for continuous wave excitation.

B. Modification of S shape via argon ion implantation

The proposed model suggests that the blue shift in the S shape of polar GaInN/GaN quantum well structures is driven by an onset of nonradiative recombination rather than by a temperature dependent occupation of states. According to this hypothesis, a significant manipulation of the S-shaped behavior should be possible by altering the radiative or the nonradiative recombination probabilities.

As a key experiment, the density of nonradiative recombination centers has been varied by a generation of argon ion implantation induced defects. With increasing implantation dose, a tremendous reduction of internal quantum efficiencies due to a severe shortening of nonradiative lifetimes has been

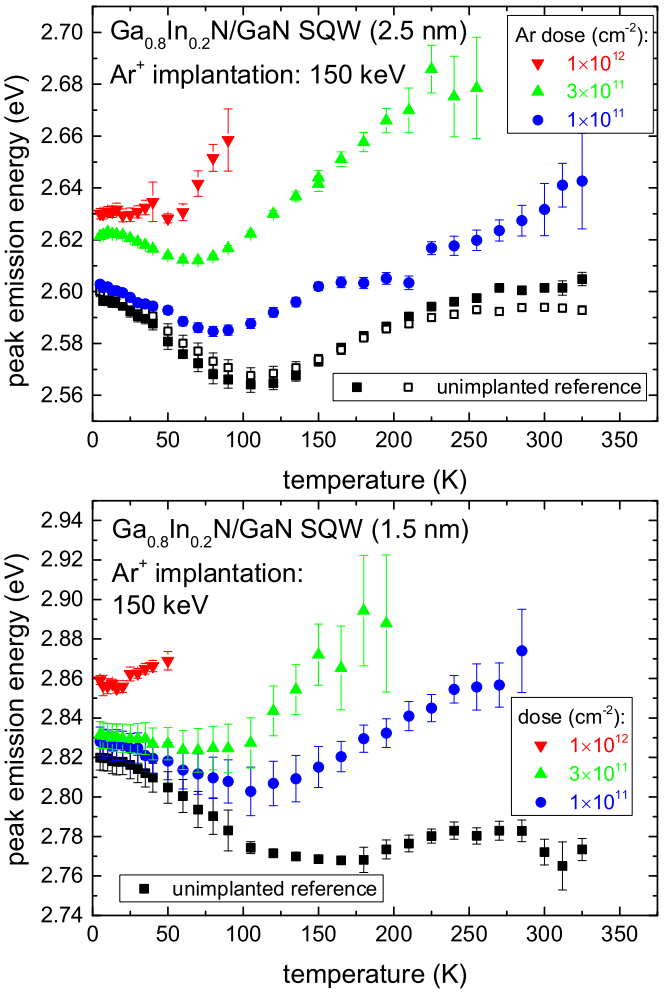

FIG. 8. (Color online) Temperature dependence of the peak emission energy of time-integrated photoluminescence spectra (pulsed excitation) before and after argon ion implantation at various doses. The shift toward higher energies sets in at lower temperatures with increasing density of nonradiative centers related to implantation induced defects. Indium compositions and quantum well thicknesses (values in brackets) are nominal values. In the upper figure, the two symbols for the unimplanted reference belong to different positions of the exitation spot at the sample. This comparison allows as an estimation of reproducibility. The implantation induced changes are significantly larger than this.

observed [26]. Figure 8 shows the temperature dependence of the peak emission energies at various implantation doses for two polar single quantum well structures with different well widths. In both cases, the onset of the blue shift occurs at reduced temperatures with increasing dose. In principle, this is in agreement with a transition between radiatively dominated and nonradiatively dominated recombination occurring at reduced temperatures after implantation. A direct comparison of intensities and blue shift as a function of temperature is rather difficult, since different nonradiative recombination channels with distinct energy dependencies may be generated during ion implantation. Yet, those implantation induced nonradiative recombination centers may differ from those being generated during the sample growth. For as-grown polar GaInN/GaN quantum wells, a coincidence of the blue shift and a drop of the intensities passing below 50% of the low-temperature values at similar temperatures is commonly observed [16,23].

According to the model of Eliseev *et al.* [15], this observation would be rather unexpected: The observed change in the onset of blue shift would imply a significant reduction of the inhomogeneous broadening after ion irradiation. A corresponding decrease of emission linewidths is not observed. Indeed, a slight blue shift at the lowest temperatures with increasing implantation dose indicate a structural change of the quantum well. Possibly, this could be the result of a material transport due to the collision cascade during implantation leading to a reduced indium content within the well. However, it is rather unlikely that the ion irradiation is beneficial for the lateral homogeneity of the quantum well.

VI. SUMMARY

We observe fundamentally different behaviors in the temperature dependence of the peak emission energy in polar and nonpolar GaInN/GaN quantum well structures; an S-shaped shift of the spectra—commonly observed for polar wells—is hardly observed for nonpolar wells on bulk GaN substrates. This indicates that the S shape is influenced by the internal piezoelectric field. Furthermore, we demonstrate a reduction of the characteristic temperature for the onset of this blue shift via argon implantation induced nonradiative recombination centers.

For polar wells, we propose an almost exponential energy dependence of radiative lifetimes caused by strongly correlated fluctuations of the transition energies and transition matrix elements due to lateral fluctuations of well width and indium composition. A thermal activation of nonradiative recombination processes likely causes a shift of the spectrum toward higher energy, if the energy dependence of the nonradiative lifetimes is weaker than of the radiative lifetimes. According to this explanation, being consistent with the experimental observations, the blue shift of the luminescence is not caused by a thermally induced change in the occupation of states, but a consequence of the competition between radiative and nonradiative recombination. Consequently, an estimation of carrier localization energies out of the S-shaped temperature dependence of peak emission energies is rather questionable.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the financial support by the Deutsche Forschungsgemeinschaft (DFG) in the frame of the Research Group FOR957 “Polarization Field Control in Nitride Light Emitters.”

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