Behavior of 2.8- and 3.2-eV photoluminescence bands in Mg-doped GaN at different temperatures and excitation densities

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The blue and ultraviolet photoluminescence bands in Mg-doped GaN have been investigated over a wide range of temperatures and excitation intensities. Redshifts of the bands were observed with increasing temperature. The bands underwent a blueshift with increased excitation density. The observed shifts of the 3.2 eV band are explained by a potential fluctuation model for a compensated semiconductor. In contrast, the shifts of the 2.8 eV band are essentially related to saturation of luminescence from distant donor-acceptor pairs responsible for this emission. Thermal quenching of the 2.8 eV luminescence band was observed at high temperatures with an activation energy of 0.3–0.4 eV. It is attributed to thermal release of trapped electrons from a deep donor state. [S0163-1829(99)02220-1]

I. INTRODUCTION

Magnesium is the preferred *p*-dopant for epitaxial GaN. Upon incorporation of Mg in GaN, a series of blue and ultraviolet emission bands are observed under optical excitation. Despite the importance of Mg, the origins of these bands are not well understood. There are three photoluminescence (PL) bands usually observed over the range of 2.5–3.3 eV. An ultraviolet (uv) band is observed with a zero phonon line at about 3.27 eV that involves donor-acceptor pair (DAP) luminescence due to optical transitions from a shallow donor to a shallow Mg acceptor.[1–3] A second PL band with a maximum near 3.1–3.2 eV is observed that arises from optical transitions from the conduction band to the same Mg acceptor (*e*-*A* transition).[3–9] In heavily Mg-doped or/and annealed *p*-type samples, a broad blue band is observed with a maximum in the range 2.5–2.95 eV,[3,5–13] whose origin is controversial. The band has been attributed to transitions from the conduction band to a deep acceptor level,[3,5,11] a Mg complex,[6,8] or DAP-type transitions from a deep donor to a shallow Mg acceptor.[7,12]

The intensity and exact positions of these broad emission bands depend on temperature[3,6,8] and on excitation density[5–8,12] which complicate their assignment. Several authors noted that a PL band with a maximum near 3.1–3.2 eV in *p*-type GaN (referred to hereafter as the 3.2 eV band) dominates at low temperature and the PL band with a maximum near 2.5–2.95 eV (referred to hereafter as the 2.8 eV band) dominates at room temperature.[6,8] The shift of the 2.8 eV blue band, with increasing excitation density, has been attributed to saturation of PL from distant DAP due to their longer lifetime,[7,12] or a change in position of the quasi-Fermi-level for holes within the impurity band.[5,6] Potential fluctuations have also been considered as a possible reason for the shift of PL bands.[5]

Since heavily Mg-doped GaN is always compensated,[7] large potential fluctuations due to randomly distributed impurities are expected. Considering that the doped material typically has no free carriers at low temperatures, the degree of potential screening should be determined by the concentration of photogenerated electrons and holes. Therefore, large potential fluctuations are expected at low excitation levels and would decrease with increasing excitation intensity.

In this work, we investigated the role of the potential fluctuations on the luminescent properties of Mg-doped GaN. PL measurements at different temperatures and excitation intensities showed that shifts of the 3.2 eV PL band in Mg-doped GaN can be explained in terms of potential fluctuations, however the large observed shift of the 2.8 eV band originates from its DAP nature.

II. POTENTIAL FLUCTUATIONS MODEL

Potential fluctuations, originating from an inhomogeneous distribution of charged defects, play a fundamental role in the optical properties of compensated and highly doped semiconductors.[14,15] In the presence of the potential fluctuations, diagonal tunnel transitions with reduced energy dominate, as shown in Fig. 1. As a result, PL bands shift to lower energy. According to the model developed by Shklovskii and Efros,[14] the typical amplitude of the potential fluctuations γ in a highly compensated *p*-type semiconductor depends on total concentration of charged donors and acceptors ($N_t = N_A^- + N_D^+$) and concentration of free poles p:

$$\gamma = \frac{e^2}{4\pi\varepsilon_0\varepsilon}\sqrt{N_t r_s}, \tag{1}$$

where r_s is the average radius of the unscreened potential fluctuations, given by

$$r_s = N_t^{1/3} p^{-2/3}. \tag{2}$$

On the other hand at very low degrees of compensation the amplitude is given by

$$\gamma = 0.26\frac{e^2}{4\pi\varepsilon_0\varepsilon}\left(\frac{4\pi}{3}N_A\right)^{1/3}\left(\frac{N_D}{N_A}\right)^{1/4} \tag{3}$$

and the screening radius in this case is given by

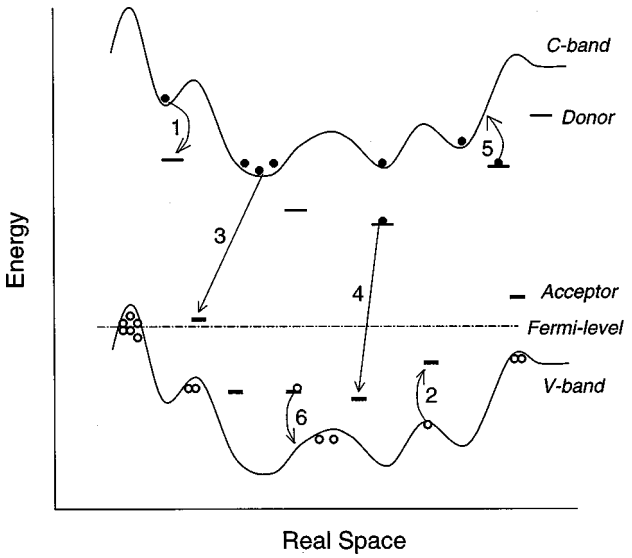

FIG. 1. Schematic representation of bands and defect levels showing the present potential fluctuations under conditions of PL. Both long- and short-range potential fluctuations are present. Arrows indicate the following transitions: 1, the capture of excess electrons by deep donors; 2, the capture of excess holes by acceptors; 3, radiative recombination of free electrons and bound holes; 4, radiative recombination in DAP; 5, the thermal release of bound electrons to the conduction band; and 6, thermal release of bound holes to the valence band.

$$r_0 = 0.58 N_D^{-1/2} N_A^{1/6}. \tag{4}$$

It follows from Eqs. (1) and (3) that potential fluctuations are larger for material with a higher concentration of ionized defects and a higher degree of compensation. For highly compensated samples, they strongly depend on the concentration of free carriers. To estimate the order of magnitude of these fluctuations, concentrations of acceptors and donors may be assumed to be 2×10^{19} and 4×10^{18} cm^{-3}, respectively, as typical values for GaN:Mg.[9] This corresponds to moderate compensation and, strictly speaking, Eqs. (1)–(4) cannot be applied. However, for qualitative analysis using Eqs. (3) and (4) one finds that $r_0 = 50$ Å and $\gamma = 10$ meV. In another limit, for a nonequilibrium carrier concentration of 10^{15} cm^{-3} (corresponding to excitation density of about 6 W/cm^2 upon assuming that the active layer width of a sample is 0.1 μm and the lifetime of free holes is 10^{-9} sec), from Eqs. (1) and (2), $r_s = 2$ μm and $\gamma = 0.7$ eV. The actual size and amplitude of potential fluctuations may be expected to be between the limiting values for a very low and very high degree of compensation.

With a decrease in excitation rate the long-range fluctuations increase dramatically according to Eq. (1). However, their amplitude cannot exceed the width of the band gap.[14] Besides, actual size and amplitude of the long-range fluctuations may be much smaller if the distribution of impurities is not random but correlated as is expected for the wide-gap semiconductors.[15] In heavily doped compensated semiconductors, the long-range potential is modulated by short-range sharp wells and humps. The size of these potential wells may be comparable to the Bohr radius of an impurity.[14]

The potential fluctuations may significantly affect the optical properties of compensated and heavily doped semiconductors. In particular, energy positions of the PL bands are redshifted due to tunnel transitions shown schematically in Fig. 1. The shifts are larger in more compensated and doped material but may be reduced by excess free carriers because of screening effects. After excitation of PL, the excess carriers are first captured by potential wells and then by an impurity and recombine. The carriers inside such wells (especially short range) are localized and each potential well resembles a single impurity center. As a result, optical transitions in heavily doped compensated semiconductors have many similar features with the DAP-type transitions. Both exhibit nonexponential luminescent time decay and PL band redshifts under a time delay. Some other features are also similar, however they have a different origin. For example, the blueshift with an increase in excitation rate for DAP emission results from saturation of PL from distant pairs emitting in the low-energy part of a spectrum. The same shift in compensated material is associated with screening of the potential fluctuations. Furthermore, with an increase in temperature, the PL from DAP may blueshift in lightly compensated material (due to thermal release of electrons from donors of distant long-lived pairs) or redshift in compensated material (due to thermal release of holes from acceptor levels of close pairs subjected to strong Coulomb interaction).[15,16] In heavily doped compensated semiconductors, increasing temperature may result in a redshift of PL bands at low excitation rates (due to thermal release of electrons or holes from the shallower potential wells) or blueshift at high excitation rates (due to increased probability of more vertical transitions).[15,17]

While the effect of potential fluctuations on the behavior of PL bands is not well understood in GaN, it has been studied in detail in a number of semiconductors including GaAs (Refs. 18 and 19) and ZnSe.[20] In these materials, for highly compensated samples with potential fluctuations, PL peaks from shallow DAP broadened and redshifted up to 120 meV from their position for uncompensated material. Increasing the excitation intensity resulted in a blueshift of the band so that at the highest excitation power, the position of the band tended to the position for uncompensated samples.[20] With an increase in temperature, the PL bands redshifted in highly compensated samples which is opposite to the shift observed for DAP emission in lightly compensated samples.[19]

III. EXPERIMENT

Epitaxial layers of Mg-doped GaN (thickness of about 2 μm) grown by metal-organic chemical vapor deposition onto *c*-plane sapphire were investigated. Highly resistive and conductive *n*- and *p*-type samples listed in Table I were examined. PL was excited with a 325 nm line of cw He-Cd laser (with power 24 mW). The excitation density was varied from 10^{-5} to 27 W/cm^2 by means of neutral density filters. The PL signal was dispersed by a 0.75 m Spex grating monochromator and detected by a Hamamatsu photomultiplier tube and a photon-counting system. The sample temperature was varied from 13 to 380 K using a closed cycle helium cryostat. A sample was pressed to the copper holder by a clip to require a good thermal contact. Heating of the illuminated area by the laser beam was estimated from a study of the

TABLE I. Characteristics of the GaN samples.

Sample no.	Mg flow rate (μmol/min)	Type	Room-temperature resistivity (Ω cm)
RK111	0	*n*	0.04
Y203	0.46	*n*	0.02
Y204	0.97	*n*	0.19
Y250	0.97	*s-i*	>100[a]
Y246	1.5	*s-i*	>100[a]
Y247	1.5	*s-i*	>100[a]
Y244	1.8	*s-i*	>100[a]
Em1,Em2,Em3		*p*	3

[a]Contacts are non-Ohmic.

variation in the exciton spectrum at different temperatures and excitation intensities in a high-purity undoped sample No. RK111. At highest excitation intensities the temperature of the illuminated area exceeded the measured temperature by about 10 K and this difference decreased with increasing temperature. All temperature-dependent data below take into account heating the sample by a laser beam.

IV. RESULTS

Typical PL spectra for four Mg-doped GaN are shown in Fig. 2. At low temperatures we have observed the broad 2.8 and 3.2 eV bands and also the shallow DAP-type PL band with a zero-phonon line at 3.25–3.29 eV.[21] The typical quantum yield was of the order of 1% for the 2.8 eV band, 10% for the 3.2 eV band, and up to 90% for the 3.25–3.29 eV band.

Low-resistivity *n*-type samples doped with Mg revealed

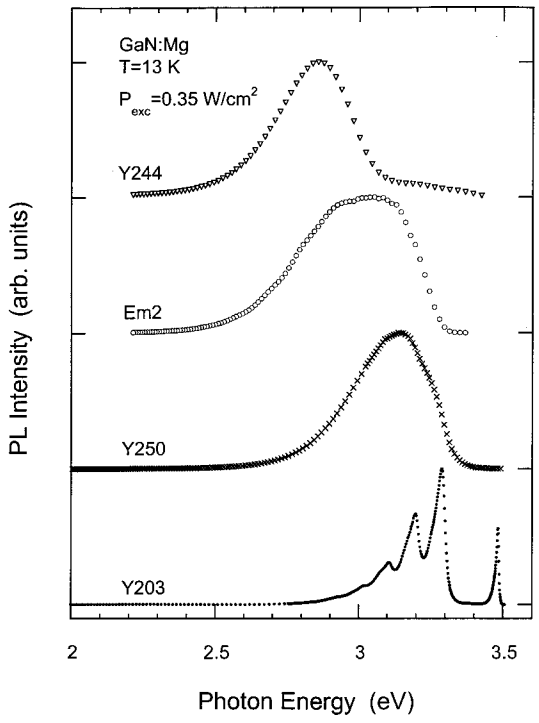

FIG. 2. PL spectra for four representative Mg-doped GaN samples.

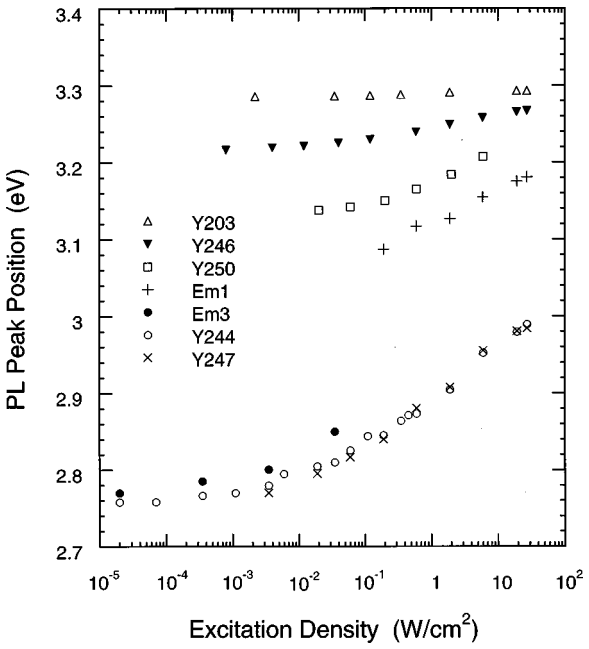

FIG. 3. Dependence of PL band maximum as a function of excitation intensities at 13 K. For samples No. Y203 and No. Y246 the shift of the zero-phonon line is shown only, whereas samples No. Em1 and No. Y250 represent the shift of the broad 3.2 eV band. Shift of the 2.8 eV band is shown for the rest of the samples.

the shallow DAP-type PL representing a zero-phonon line at about 3.29 eV and several LO-phonon replicas as shown in Fig. 2. The position of a zero phonon peak and its phonon replicas were essentially independent of excitation power; Fig. 3 shows the change in position for the zero-phonon line only. In contrast, in semi-insulating and *p*-type samples, positions of the 2.8 and 3.2 bands depended strongly on excitation intensity (Fig. 3). Figure 4 shows the PL spectrum of a sample where the 2.8 eV band is dominant. A large continuous blueshift of the 2.8 eV band with increasing excitation power was observed for all samples revealing this band. Under high excitation conditions, the total shift exceeds 0.2 eV. The shape of the band is nearly independent of excitation power and the full width at half maximum (FWHM) of this band is about 300–350 meV.

In low-resistivity *p*-type samples (Nos. Em1, Em2, and Em3), the 2.8 eV band overlapped the 3.2 eV band (shown in Fig. 2) and its study was possible only at a low excitation level. With increasing excitation intensity, the intensity of the 2.8 eV band saturates at an excitation density (P_{exc}) of over 0.1 W/cm^2 and only the 3.2 eV band is observed at high excitation levels. In the samples where the 2.8 eV band predominates (Nos. Y244 and Y247), its intensity was linearly dependent on excitation density up to 27 W/cm^2. Time-resolved PL measurements showed that at low temperature the lifetime of the 2.8 eV PL is about 10^{-5} sec and the decay is nonexponential.

A substantial shift of the 3.2 eV band with excitation power has also been observed in several samples. In sample No. Em1, an increase of excitation power by 1000 resulted in a blueshift of the band maximum of 0.1 eV from 3.1 to 3.2 eV (Fig. 5). Likewise, high-resistivity samples with a predominant 3.2 eV band also revealed a significant blueshift with increased excitation power (Fig. 3).

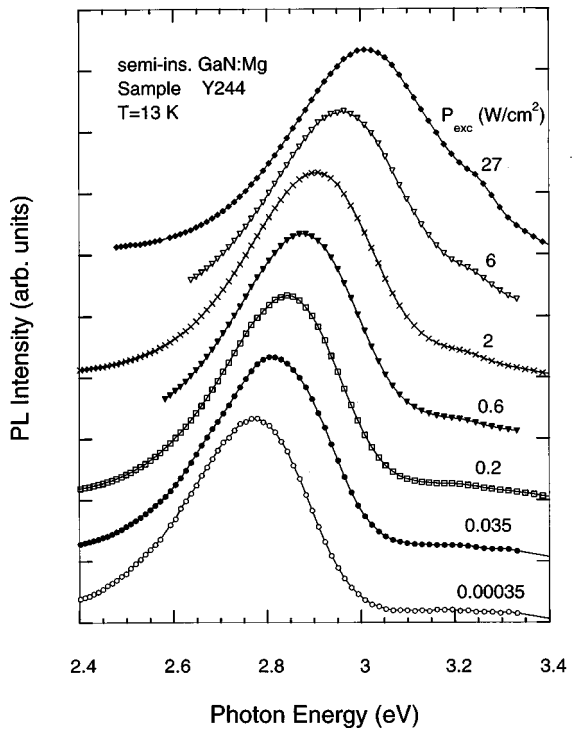

FIG. 4. PL spectra for sample No. Y244 at different excitation intensities. The shape of the 2.8 eV PL band is independent of excitation intensity except at the highest excitation density (the spectrum at 27 W/cm^2 is slightly broadened and blueshifted due to visible overlap with the 3.2 eV band).

In some samples, the broadband at 3.2 eV gradually transformed into PL spectra with a peak at 3.27 eV with characteristic phonon replicas, typical for lightly Mg-doped GaN (Refs. 1–3) (Figs. 6 and 7). The observed transformation

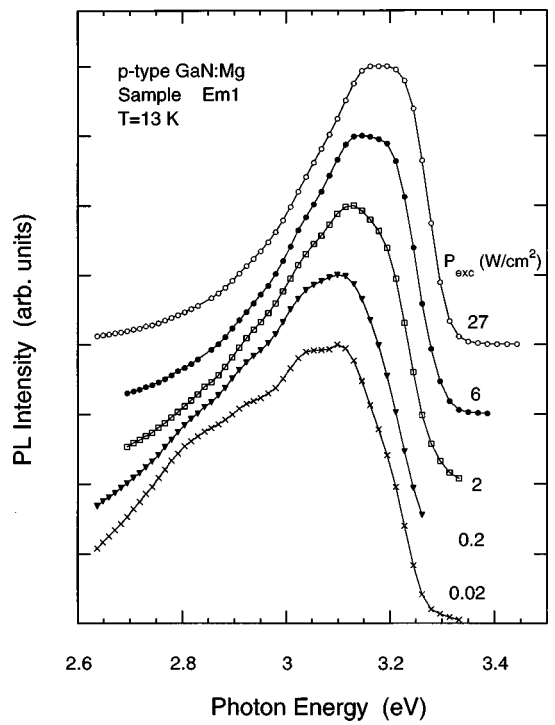

FIG. 5. PL spectra for sample No. Em1 at different excitation intensities.

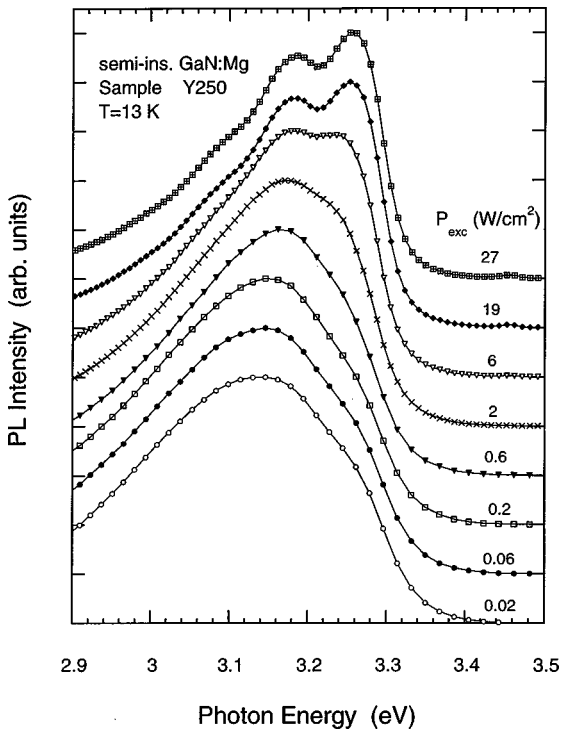

FIG. 6. PL spectra for sample No. Y250 at different excitation intensities.

may be explained by the reduction of potential fluctuations as a result of screening from the photoexcited carriers. This indicates that the 3.27 eV band and a broad structureless band at 3.1–3.2 eV have the same origin. Their differences in shape and position are due only to potential fluctuations.

An additional broadband was occasionally noted as a

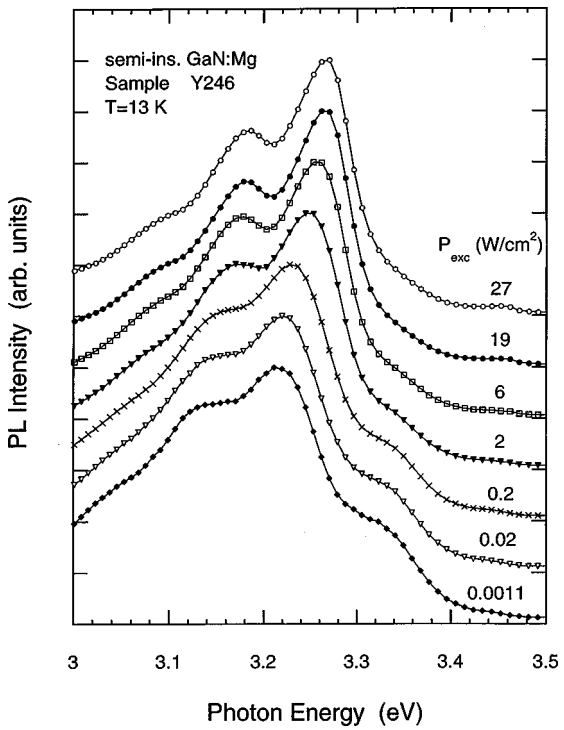

FIG. 7. PL spectra for sample No. Y246 at different excitation intensities.

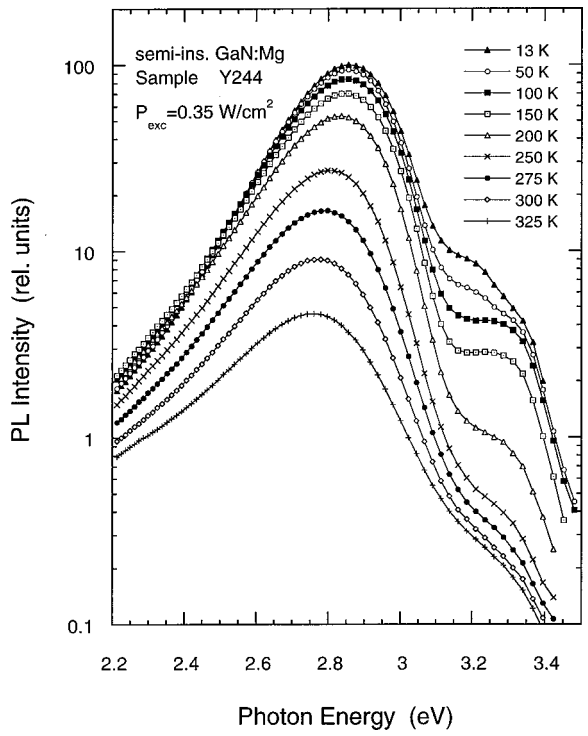

FIG. 8. PL spectra for sample No. Y244 at different temperatures.

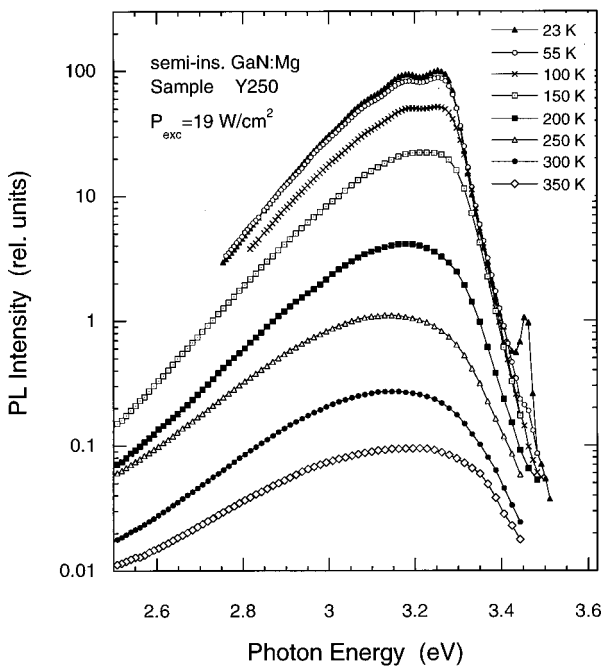

FIG. 9. PL spectra for sample No. Y250 at different temperatures.

shoulder with a maximum at about 3.3 eV (Fig. 7). The origin of this band is unknown and it apparently dominates over the similar 3.2 eV band at elevated temperatures, which can lead to misinterpretation of the PL spectrum at room temperature.

In addition to the aforementioned PL bands, several samples revealed a comparatively sharp band at a photon energy of 3.45–3.46 eV. The intensity of this band, which is most probably due to an exciton bound to a neutral acceptor, increased quadratically with an increase in excitation intensity, whereas the dependence of the 2.8 and 3.2 eV bands was nearly linear over the entire range of excitation densities for almost all samples.

The variations in the PL spectra due to temperature were studied and are shown for two representative samples. In Fig. 8, the 2.8 eV band dominates at room temperature and the 3.2 eV band appears at low temperature as a shoulder (an additional peak, seen at $T=50-150$ K, is most probably the aforementioned 3.3 eV band). With increasing temperature, the FWHM of the 2.8 eV band was roughly invariant up to 200–250 K. In the sample shown in Fig. 9, the 3.2 eV band dominates over the entire temperature range.

The 2.8 and 3.2 eV bands in several high-resistivity and conductive *p*-type samples redshifted with increasing temperature and this shift, particularly striking at low temperature, depended on excitation density. The 2.8 eV band redshifted 20–40 meV up to 100 K at high excitation density but its position remained nearly temperature independent at low excitation density (Fig. 10). This behavior may be explained by assuming that this band has a deep DAP character. In this case, the closest pairs contribute to the high-energy portion of the spectrum and at a high excitation rate this contribution increases because of saturation of PL from distant pairs. The Coulomb interaction between close pairs may be very strong[22] and the resultant ionization energy will decrease sufficiently for thermal release of the trapped carriers. As a result, the high-energy portion of the spectrum begins quenching at lower temperatures and the PL band undergoes a redshift.[15] However at a low excitation rate, PL from comparatively distant pairs dominates and the redshift

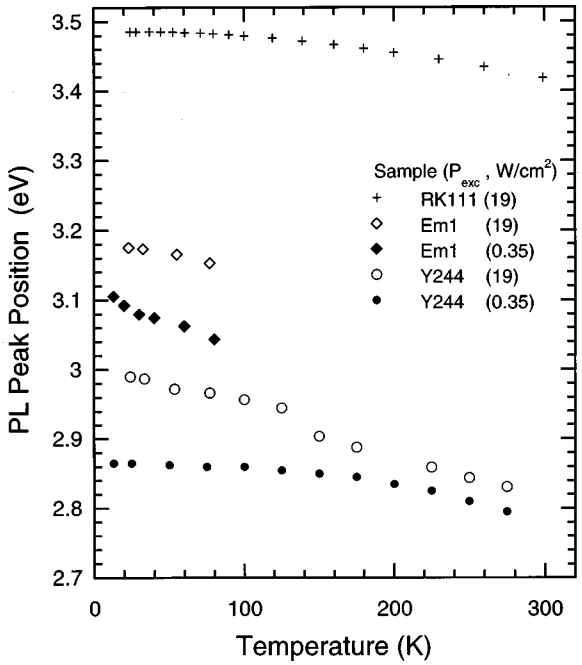

FIG. 10. Shifts of the PL band peaks with temperature under different excitation intensities ($P_{exc}=0.35$ and 19 W/cm^2). Sample No. Y244 represents the 2.8 eV band; No. Em1 represents the 3.2 eV band. Temperature dependence for a free exciton peak is also shown for a pure undoped sample No. RK111 to demonstrate the variation of the gap width in GaN.

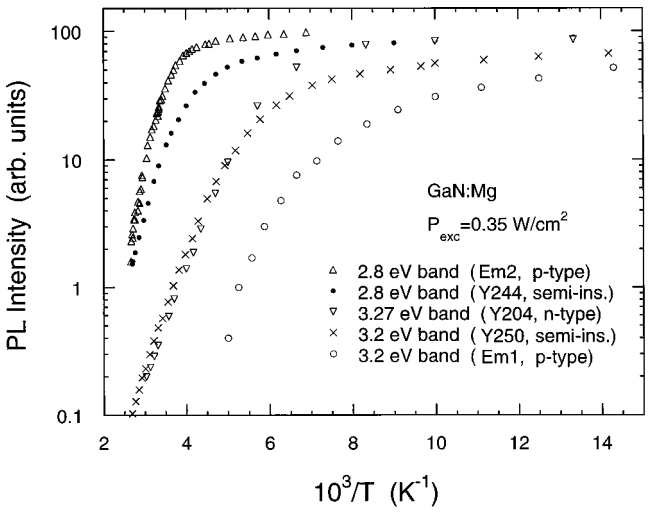

FIG. 11. Temperature dependence of PL intensity for different GaN:Mg samples.

is reduced. This effect has been previously observed in Si doped with donors and acceptors having similar activation energies.[16] The observed temperature-induced redshift at high excitation rate is just the reverse of the well-known blueshift of the DAP emission involving shallow donors and acceptors. In the latter case the blueshift is due to thermal escape of electrons from the shallow donor which is more favorable for the long-lived distant pairs contributing to the low-energy part of a spectrum. The observed redshift of the 2.8 eV band with an increase in temperature cannot be attributed to the potential fluctuations since the redshift caused by the potential fluctuations should increase with a decrease of excitation rate.[15]

In contrast, the 3.2 eV band, at least in sample No. Em1, had a large redshift with temperature that increased with decreasing excitation rate as shown in Fig. 10. This behavior may be explained in terms of the potential fluctuation model whereby there is a thermal release of free carriers from the shallower short-range wells and their percolation into the deeper ones.[15] This effect has been previously observed in compensated Ge (Ref. 17) and GaAs.[19]

In Fig. 11, the logarithmic PL intensity as a function of inverse temperature is plotted. With increasing temperature, both the 2.8 and 3.2 eV bands were thermally quenched. The quenching is thermally activated. The intensity of the 3.2 eV band in both highly resistive and conductive *p*-type samples decreased with temperature even at the lowest temperatures. The nominal activation energy increased with increasing temperature up to about 200 meV. In contrast, in *n*-type samples, the intensity was independent of temperature up to $T=100$ K and subsequently decreased with an activation energy of 200 meV. The quenching is attributed to thermalization of trapped holes to the valence band. As the intensity of the 3.2 eV band decreased with temperature, an increase in the near-interband PL at 3.46 eV was noted in several samples. This observation supports the assertion that the temperature quenching of the 3.2 eV band above 100 K results from thermalization of trapped holes from the Mg acceptor into the valence band. The continuous decrease of PL intensity with temperature in the semi-insulating and *p*-type samples may be similarly explained. However, in this case there is a distribution of activation energies for different acceptors as a result of short-range potential fluctuations as well as Coulomb interaction with compensating donors in these samples.[15] The thermal quenching of the 3.2 eV PL band in low-resistivity *p*-type samples is not as evident, since according to our estimates, at these temperatures the concentration of neutral acceptors considerably exceeds the concentration of charged acceptors and greatly exceeds the concentration of free holes. The observation of quenching in this case may support potential fluctuations, since the luminescence is mainly governed by the recombination of free electrons from potential wells and the probability of acceptors being filled by electrons in equilibrium is higher in the vicinity of such wells (Fig. 1).

In the case of the 2.8 eV band, the intensity was weakly dependent on temperature below 200 K and at high excitation rate the decrease of the integrated PL intensity was more pronounced due to fast quenching of the high-energy part of the spectrum. At temperatures above 250 K, a sharp decrease in intensity of this PL has been observed with an activation energy of about 0.3–0.4 eV as shown in Fig. 11.

V. DISCUSSION

The shift of a PL band with excitation intensity may arise from several different processes. Among the most probable are (i) potential fluctuations in compensated samples, (ii) changes in the DAP-type emission due to differences in the lifetimes of close and distant pairs, and (iii) impurity band formation.

Formation of a Mg impurity band may be ignored for concentrations of less than $\sim 10^{20}$ cm^{-3} (corresponding to the Mott transition for the Mg acceptor with a Bohr radius of 5 Å). In more heavily doped material the potential fluctuations are expected to play a more important role than the impurity band formation.[14]

We propose that the 3.2 eV PL band in GaN:Mg involves a free electron to bound acceptor transition and the observed shifts result from potential fluctuations. The large blueshift of this band in semi-insulating and *p*-type samples cannot be explained by saturation of PL from distant pairs with an increase of excitation intensity. Indeed, the blueshift for DAP-type transitions should be very small in the case of shallow donors and a high concentration of acceptors.[15] In addition, optical transitions most probably take place from the conduction band to acceptor level in the samples with a high concentration of donors and acceptors.[15]

The explanation for large blueshifts of the 2.8 eV band with variation of excitation intensity depends on the assignment of this band. It is known that the 2.8 eV band appears only at high Mg doping concentrations (over 10^{19} cm^{-3}) in GaN.[2,7] It has been theoretically predicted that at high acceptor concentrations the formation of native donor defects occurs.[23] It was recently suggested that the 2.8 eV band is due to optical transitions from the deep donor (350–430 meV) to a shallow Mg acceptor.[7,12]

Let us consider the deep DAP model for this PL band, as applied to the case of a highly doped compensated semiconductor. This PL band appears usually at high concentrations of Mg (10^{19}–10^{20} cm^{-3}),[2,7] which corresponds to a mean separation between impurities of about 15–30 Å. In spite of

the high localization of the wave functions for electrons and holes bound to the defects (the Bohr radius of about 7 and 5 Å, respectively, in the effective-mass approximation), the probability of such interimpurity transitions may be quite high.[22] Note in the case under consideration the Coulomb interaction between charged donors and acceptors may be very large and, hence, the uncharged donor would have a much deeper level than may be found from the PL measurements. Besides, one may expect the wide scatter in the Coulomb energies for different pairs contributing to the PL band.[22]

The long-range potential fluctuations should not significantly affect the deep DAP emission because of small separations in pairs contributing to PL. These transitions may be considered as almost vertical in the scale of long-range fluctuations. Therefore, the role of the potential fluctuations is reduced. Consequently, the large blueshift of the PL band with increasing excitation rate can only be explained by differences in the radiative lifetimes of close and more distant pairs. Very similar blueshifts of the 2.8 eV band with variation of excitation intensity, observed for several samples (Fig. 3), also point to the suppression of the effect of potential fluctuations on the deep DAP emission, as well as the explanation for the redshift of this band with temperature discussed above.

The position of the 2.8 eV band at room temperature and low excitation rate (2.75 eV in our experiments) suggests the optical position of a deep donor at about 0.50 eV below the conduction band. This compares to a measured value of 0.3–0.4 eV in this study found from thermal quenching experiments at $T>200$ K. The difference may be attributed to a Stokes shift. The shape of the band also suggests strong electron-phonon interaction with possible atomic relaxation between the ground and excited states of the defect. It should be noted that the magnitude of donor energy is underestimated because of neglecting the Coulomb interaction between charged donors and acceptors.

The values of the radiative lifetime for the 2.8 eV band found in this study (about 10 μsec) and by Seitz *et al.*[13] (about 0.2 msec) are consistent with DAP emission. Similar lifetimes were observed for the Frenkel pairs in ZnSe (about 0.1 msec).[22] (In the latter case the Bohr radius of deep defects was estimated as 2.0 and 2.4 Å, and the separations between most of the ODMR pairs were in the range 10–20 Å.)

VI. CONCLUSIONS

Photoluminescence of *n*- and *p*-type and also semi-insulating GaN deliberately doped with Mg has been studied over a wide range of temperatures and excitation intensities. Two broad PL bands (blue and uv) are observed with nominal peaks at 2.8 and 3.2 eV. Large redshifts of the bands with temperature were observed. For the 2.8 eV band, the shift is associated with the DAP nature, whereas for the 3.2 eV band, the shift is attributed to the presence of potential fluctuations. The uv band, having a maximum in the range 3.1–3.2 eV at low temperature, undergoes a blueshift with an increase of excitation intensity and gradually transforms into a PL band with several phonon replicas, typical for lightly Mg-doped GaN. These findings indicate that a structureless band with a maximum near 3.1–3.2 eV in moderately and heavily Mg-doped GaN involves the same free to bound transition to shallow acceptors as that observed in lightly Mg-doped GaN. Their differences in shape and position result from potential fluctuations.

The observed blueshift of the 2.8 eV band from 2.7 to 3.0 eV with increasing excitation rate is mostly due to differences in the Coulomb interaction for close and more distant pairs. This band is attributed to transitions from a deep donor to the shallow acceptor Mg. Thermal quenching of this band begins at $T>200$ K with an apparent activation energy of about 0.4 eV. The observed quenching is attributed to the thermalization of trapped electrons from the deep donor state to the conduction band.

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