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# Neutral-donor-bound exciton recombination dynamics in GaN grown by metalorganic chemical vapor deposition

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Neutral-donor-bound exciton recombination ( $I_2$ ) dynamics have been studied by photoluminescence in an unintentionally doped  $n$ -type GaN epitaxial layer grown by metalorganic chemical vapor deposition. The luminescence emission line shape, peak position, and intensity as functions of temperature have been measured. In particular, time-resolved emission spectroscopy has been employed to study the dynamic processes of the bound exciton recombination, from which the temperature and the emission energy dependencies of the recombination lifetime of this transition have been obtained. © 1995 American Institute of Physics.

GaN has been recognized as one of the most important semiconductors recently<sup>1-5</sup> due to its potential for many device applications, including UV-blue light emitting diodes (LED) and laser diodes. There has been a considerable amount of research effort directed towards the understanding of its optical properties. However, the investigation and understanding of its fundamental band-edge transitions, including the band-to-band, exciton, and band-to-impurity transitions, are still in their infancy due to the lack of high-quality crystals. In particular, studies of excitonic transitions in GaN are extremely important for the understanding of fundamental properties of this material as well as for its practical applications.<sup>6</sup> Most importantly, the dynamics of exciton transitions can provide important information regarding excitation and energy transformation processes and recombination lifetimes, which are strongly correlated with quantities such as the quantum efficiency and optical gain in GaN. These basic quantities are crucial to the design of optoelectronic devices.

In this letter, we report an experimental investigation on the neutral-donor-bound exciton recombination ( $I_2$ ) dynamics in an unintentionally doped  $n$ -type GaN. The temperature dependencies of the luminescence linewidth, emission intensity, and peak position have been measured. Time-resolved luminescence emission spectroscopy has been employed to study the dynamical behavior of the  $I_2$  transition. The recombination lifetimes at different temperatures and emission energies have been measured, from which the radiative recombination lifetime has been obtained.

The GaN sample used here was grown using low-pressure metalorganic chemical vapor deposition (MOCVD). Prior to the deposition of the GaN layer a thin 50 nm AlN buffer layer was grown on the sapphire ( $\text{Al}_2\text{O}_3$ ) substrate. A room-temperature electron concentration (due to unintentional doping) of about  $2.4 \times 10^{17} \text{ cm}^{-3}$  was determined by Hall measurements. The sample was held inside a closed-cycle He refrigerator. Photoluminescence spectra were col-

lected in a reflecting mode. An excitation pulse of about 7 ps at a repetition rate of 9.5 MHz was provided by a cavity-pumped dye laser, which was pumped by an yttrium-aluminum-garnet (YAG) laser with a frequency doubler. The output from the dye laser was frequency doubled again by a second frequency doubler to provide tunability in the UV region. A single photon counting detection system was used to record the time-resolved photoluminescence spectra. With the use of a deconvolution technique, the recombination lifetime can be measured down to 50 ps.

Figure 1 shows the continuous-wave (cw) spectrum of the GaN sample measured at  $T = 10 \text{ K}$ . Two transition peaks near 3.476 and 3.483 eV can be clearly resolved. The transition line at 3.476 eV is due to the recombination of the

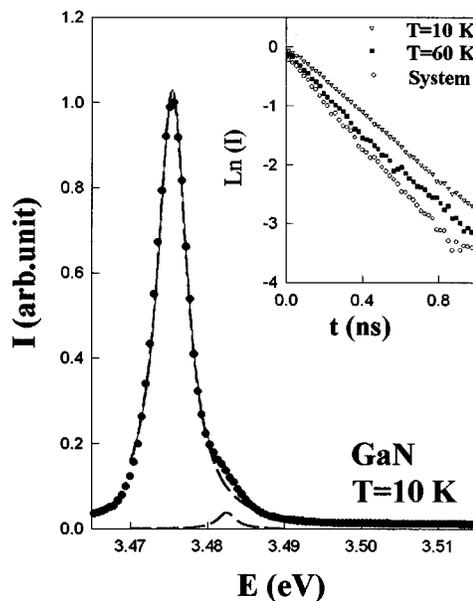


FIG. 1. Continuous wave photoluminescence spectra of an unintentionally doped  $n$ -type GaN measured at  $T = 10 \text{ K}$ . The solid line is the least-squares fit of data to the Lorentzian function of Eq. (1) for two individual peaks (the dotted lines). The inset shows the semilogarithmic plots of the temporal responses of the excitonic transition measured at the  $I_2$  peak positions for two representative temperatures at  $T = 10$  and  $60 \text{ K}$ . The instrumental response to laser pulses (7 ps width) is indicated as "system."

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excitons bound to neutral donors associated with nitrogen vacancies ( $D^0$ ,  $X$ ) or the  $I_2$  line.<sup>2,3</sup> The transition line at 3.483 eV is due to the recombination of free excitons (FX).<sup>2</sup> These results are consistent with previous measurements on these transitions.<sup>7-9</sup> The emission lines can be fitted quite well (except in the low-energy tail region) by a Lorentzian function,

$$I(E) = \sum_{i=1}^2 \frac{2A_i}{\pi} \frac{\Gamma_i}{4(E-E_i)^2 + \Gamma_i^2}, \quad (1)$$

as shown as the solid line in Fig. 1. The dotted lines are the fitted two individual peaks of the  $I_2$  and FX emission lines. In Eq. (1),  $A_i$ ,  $\Gamma_i$ , and  $E_i$  ( $i=1$  and  $2$ ) are the luminescence peak intensities, the full widths at half-maximum (FWHM), and peak positions of  $I_2$  ( $i=1$ ) and FX ( $i=2$ ) transitions, respectively. The fitted values are  $E_1=3.4754$  eV,  $E_2=3.4825$  eV,  $\Gamma_1=4.6$  meV, and  $\Gamma_2=2.9$  meV from the  $T=10$  K spectrum. From these measurements, we obtain a value of 7.1 meV for the binding energy of the neutral-donor-bound exciton,  $E_{\text{bx}}$ . The intensity ratio of the FX to the  $I_2$  emission lines increases with an increase of temperature and excitation intensity as expected.<sup>10</sup> The small  $\Gamma$  values indicate that the sample under investigation is of high crystalline quality.

The line shapes of the free- and bound-exciton transitions have been well studied in other semiconductors.<sup>11</sup> It has been pointed out that<sup>11</sup> if the linewidth broadening is caused predominantly by crystal imperfection, impurities, and strain, then a Gaussian line shape is expected; if the linewidth broadening is caused primarily by exciton-phonon interactions, a Gaussian line shape is expected for a strong exciton-phonon interaction and a Lorentzian line shape for a weak exciton-phonon interaction.<sup>11</sup> We have employed both Gaussian and Lorentzian line shapes to fit the emission spectra. However, the emission spectra obtained at different temperatures can only be fitted by the Lorentzian function of Eq. (1), which indicates that weak exciton-phonon interaction is a prevailing process in GaN, similar to the case in other III-V compounds.<sup>11</sup>

The inset of Fig. 1 shows semilogarithmic plots of the temporal responses of the  $I_2$  transition measured at the peak positions for two representative temperatures of  $T=10$  K and 60 K. The decay of the  $I_2$  transition is single exponential,  $I(t)=I_0 \exp(-t/\tau)$ , where  $\tau$  defines the recombination lifetime. By comparing the temporal responses of the  $I_2$  transition at these two temperatures, we see that luminescence decays faster at 60 K than at 10 K.

In Fig. 2 we present the time-resolved emission spectra obtained at  $T=10$  K for several representative delay times. Unlike the band-to-impurity transition in GaN where the peak position shifts to lower energies as the delay time increases,<sup>12</sup> the peak position of the  $I_2$  transition is almost independent of delay times. This is consistent with the observation (shown in Fig. 4) that the variation of the recombination lifetime of the  $I_2$  transition with emission energy ( $d\tau/dE$ ), is much smaller than that of the band-to-impurity transition.<sup>12</sup> The free-exciton transition is very difficult to resolve in the time-resolved spectra shown in Fig. 2 due to

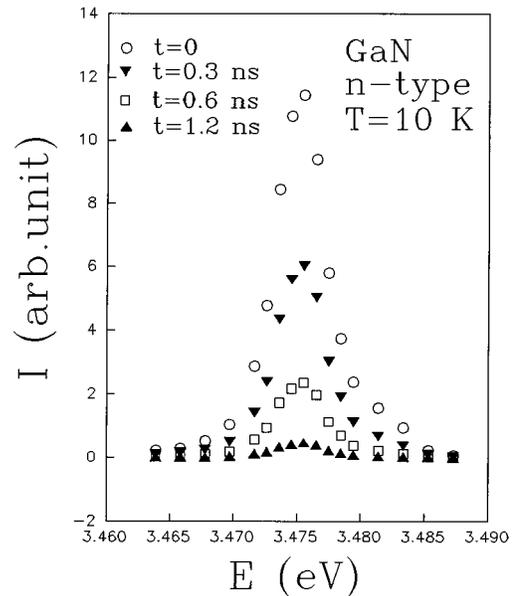


FIG. 2. Time-resolved emission spectra measured at  $T=10$  K for several representative delay times.

its relatively weak signal at 10 K and low-energy resolution of the spectra.

In Fig. 3, we plot (a) the peak position  $E_p$ , (b) the (FWHM), and (c) the emission intensity of the  $I_2$  transition as functions of temperature. Figure 3(a) shows that the  $I_2$  transition follows the energy gap variation with temperature as expected. The FWHM shows a strong dependence on temperature, but simple linear relation with temperature<sup>11</sup> expected from acoustic phonon-exciton interaction is not observed.

Figure 3(c) shows the thermal quenching of the  $I_2$  transition. The temperature dependence of the emission intensity of the  $I_2$  transition,  $I(T)$ , can be described by

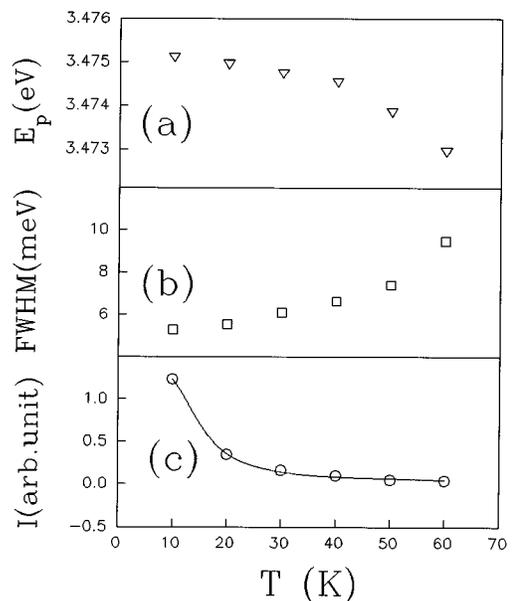


FIG. 3. Temperature dependence of the (a) peak position  $E_p$ , (b) FWHM, and (c) emission intensity of the  $I_2$  transition. The solid line in (c) is the least-squares fit of data by Eq. (2).

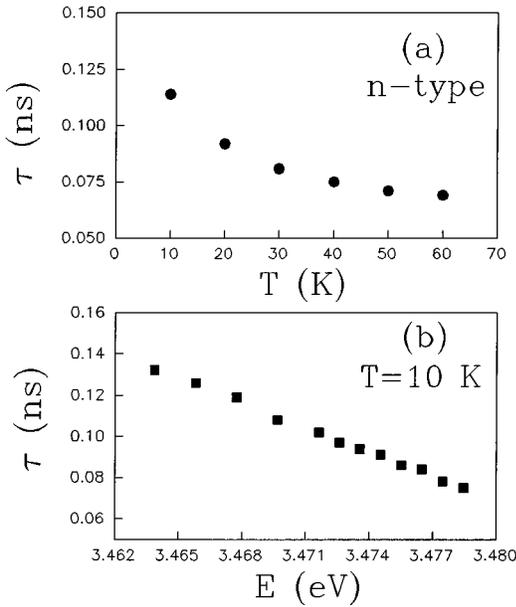


FIG. 4. (a) Temperature dependence of the  $I_2$  recombination lifetime measured at the luminescence spectral peak positions. (b) Emission energy dependence of the  $I_2$  recombination lifetime measured at  $T=10$  K.

$$I(T) = \frac{I_0}{1 + C \exp(-E_0/kT)}, \quad (2)$$

where  $I_0$  is the emission intensity at  $T=0$ ,  $C$  is a constant, and  $E_0$  is the activation or dissociation energy of the neutral-donor-bound excitons. We have used Eq. (2) to fit our experimental data (the open circles) shown by the solid line in Fig. 3(c). The fitted values are  $C=8.2$  and  $E_0=5.9$  meV. The fitted value of  $E_0=5.9$  meV is very close to the binding energy of neutral-donor-bound exciton,  $E_{bx}$ , which is about 7.1 meV from Fig. 1. It thus indicates that the thermal quenching of  $I_2$  transition is due to the dissociation of neutral-donor-bound exciton described by



where  $D^0$  and  $X$  are the neutral donor and free exciton, respectively. This conclusion is consistent with our observation that the intensity ratio of the free exciton to the  $I_2$  transition increases with an increase of temperature. The slight difference between the fitted value of  $E_0$  and  $E_{bx}$  may be due to the broadening of the  $I_2$  transition and the process of bound-exciton dissociation into other recombination channels at higher temperatures.

The temperature dependence of the recombination lifetime of the  $I_2$  transition measured at its spectral peak positions is shown in Fig. 4(a). The  $I_2$  transition decays very fast with a lifetime of about 100 ps at low temperatures. Figure 4(a) shows that  $\tau$  decreases with increasing temperature. This behavior is due to the increased nonradiative recombination rate at higher temperatures, related with the dissociation of the neutral-donor-bound excitons as described by Eq. (3) or into other recombination channels. The radiative recombination lifetime of the  $I_2$  transition can be obtained by extrapolating  $\tau$  to  $T=0$ , which is about 130 ps. At temperatures  $T$

$>60$  K, the  $I_2$  recombination lifetimes cannot be measured due to the limitation of time resolution of our current detection system.

Figure 4(b) shows the emission energy dependence of the recombination lifetime of the  $I_2$  transition measured at  $T=10$  K, which shows  $\tau$  decreasing with increasing emission energy. It has been predicated and confirmed in many materials that the radiative recombination lifetime of the bound exciton increases with an increase of its binding energy,  $E_{bx}$ .<sup>13</sup> The linewidth of the  $I_2$  emission band indicates the existence of a distribution of  $E_{bx}$  in the investigated sample with higher emission energies corresponding to lower values of  $E_{bx}$ . Thus  $\tau$  decreases with an increase of emission energy.

The observed radiative recombination lifetimes of about 130 ps for the  $I_2$  emission line in GaN is about a factor 4 shorter than that of the  $I_2$  transitions in better studied semiconductors such as CdS.<sup>14</sup> The radiative quantum efficiency  $\eta$  can be written as<sup>10</sup>

$$\eta = [1 + (\tau_r/\tau_n)]^{-1}, \quad (4)$$

where  $\tau_r$  and  $\tau_n$  are the radiative and nonradiative decay lifetimes of optical transitions. Thus the shorter the radiative recombination lifetime is, the higher the quantum efficiency. The implications of short radiative recombination lifetime of the  $I_2$  transition observed in GaN indicates that UV-blue optical detectors and laser diodes with higher quantum efficiencies  $\eta$  and faster responses can be obtained by using GaN.

In conclusion, we have studied neutral-donor-bound exciton recombination dynamics in a MOCVD grown  $n$ -type GaN epitaxial layer. The luminescence linewidth, peak position, and emission intensity as functions of temperature have been measured, from which information concerning exciton-phonon interaction as well as dissociation process of neutral-donor-bound exciton have been obtained. Time-resolved emission spectra of the  $I_2$  transition have been measured. Recombination lifetimes as well as their temperature and emission energy dependencies have also been determined.

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