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Impact ionization of excitons in an electric field in GaN

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Abstract
The impact ionization of excitonic states is studied in epitaxial GaN films and GaN/AlGaN quantum well structures. The investigation is carried out by optical methods involving the observation of quenching of the exciton photoluminescence under an applied electric field. It is found that impurity scattering rules the momentum and energy relaxation, rather than the acoustic phonon scattering. The effective mean free path of hot electrons is estimated. In GaN/AlGaN quantum wells the mean free paths of hot electrons appear to be an order of magnitude larger than those of GaN films due to the decrease in scattering probability of the electron in the two-dimensional case.

1. Introduction

In recent years much attention has been devoted to the study of III nitride compounds as perspective wide band gap semiconductor materials for designing modern optoelectronic devices that could operate in both the visible and in the ultra-violet spectral region [1]. Nevertheless, many fundamental properties of these materials are not known in detail. In particular, few efforts have been directed to the study of the optical properties of GaN and related materials under an external electric field. Until recently, there were only a few papers dealing with the influence of external electric field on the excitonic states in III nitrides [2, 3]. At the same time, such investigations seem to be very important. The radiative recombination, taking place, at least at low temperatures, through the excitonic states, coexists in optoelectronic devices with the current going through the structure. Therefore, the comprehension of the interaction between charged carriers and excitons becomes essential.

The work we present in that communication is devoted to the investigation of impact ionization (or delocalization) of excitons in gallium nitride. The investigation is carried out by optical methods involving the observation of quenching of the exciton photoluminescence

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under applied electric field. The quenching is due to disintegration of excitons or exciton complexes by hot carriers that have acquired the necessary energy from an electric field. The number of such high-energy electrons depends strongly on the mechanisms that rule their energy and momentum relaxation. Therefore, the study of the quenching effect in dependence on electric field can provide valuable information about these mechanisms. The advantage of optical methods is that it allows us to study the impact ionization of weakly bound states and investigate separately the states with different ionization energy. Besides the basic physics phenomena that are investigated, our study is made on a series of epilayers grown by different methods: ELOG-MOVPE, MOVPE and MBE. The effect of impact ionization was also studied in MBE-grown GaN/AlGaN quantum well (QW) structures. It is found that impurity scattering rule the momentum and energy relaxation, rather than the acoustic phonon scattering.

2. Experiment

The samples under study were residual n-type GaN films grown on c-plane sapphire substrates. The ELOG-GaN sample used is this study was grown by MOVPE at atmospheric pressure using a two step process [4]. Typically, a 10 μm periodic grating of 3–5 μm wide stripes, aligned along $\langle 10-10 \rangle$GaN, are opened in an Si_xN_y mask deposited on a GaN template layer. Then the GaN growth proceeds until full coalescence is achieved. NH_3 and solid sources for group-III elements were used for the MBE growth of the nitride layers and GaN/AlGaN quantum wells. After a rapid nitridation step, a 250 Å thick GaN buffer layer is deposited at 500 °C and further annealed at 900 °C before growing the GaN epilayer at 800 °C (for more details concerning the growth conditions see [5]). The GaN/AlGaN QW structures consist of $Al_{0.9}Ga_{0.91}N$ barriers and GaN single quantum well with the thickness of some monolayers (ML). The ohmic contacts were formed by vacuum deposition of Al electrodes (two parallel strips of some millimetres in length with a gap between contacts of 0.2–0.3 mm) as shown in figure 1. The 325 nm line of an He–Cd laser was used to excite the photoluminescence. The laser beam was focused into a spot about 0.5 mm in diameter covering the whole distance between the electrodes. During the photoluminescence experiments the sample was immersed into liquid helium. A dc voltage was applied to the structure. When this voltage was applied, the current (of a few mA) went through the sample. It could produce sample heating. To ensure

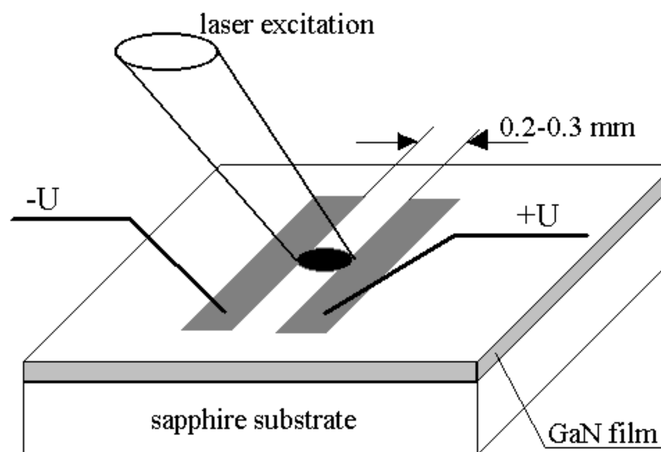

Figure 1. Structures under investigation.

the best cooling conditions, the liquid helium was pumped down and the measurements were made at the temperature ($T = 2$ K) below the λ-point, the temperature at which the liquid helium undergoes the transition to a super-fluid state. In this case there is no vapour interlayer between the sample surface and the cooling agent, as it take place when the helium temperature is above the λ-point. In the last case ($T = 4.2$ K) the vapour interlayer worsens the heat removal from the sample and a rather small current can produce significant sample heating. The tests, which were made during the experiments, have shown that the sample heating was insignificant and had thus poor influence on the luminescence spectra. In particular, the quenching of the exciton photoluminescence of a GaN/AlGaN quantum well under applied voltage was studied at two temperatures: $T = 2$ K and $T = 4.2$ K. The dependences of the luminescence intensity on the voltage were completely the same at both temperatures, indicating that we really deal with the electric field effect because the thermal quenching should be much more pronounced at $T = 4.2$ K due to the worse cooling conditions.

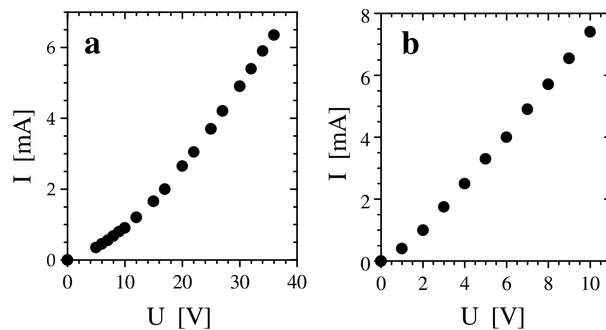

Figure 2. I–V characteristics of two samples under investigation: (a) GaN film on sapphire substrate; (b) 17 ML thick GaN/AlGaN quantum well.

During the photoluminescence experiments the current going through the sample was monitored and then the current–voltage (I–V) characteristics were plotted. Two of them are presented in figure 2. The I–V characteristics of the samples were more or less linear. In some samples the I–V characteristics have a nonlinear part at small voltage, but at higher voltage it becomes relatively linear. So, we are able to consider the electric field in a sample as homogeneous, at least at not so small voltage.

3. Results

An intense line (exciton bound to a neutral donor) dominates in the luminescence spectra of our samples in the $A_{n=1}$-exciton resonance region. The much weaker free exciton line [6] can also be distinctly seen in the spectra. In some samples not only one, but two bound exciton lines with different binding energies were observed. The BE and BE1 transitions are the usual residual neutral donor bound exciton line in GaN. We attribute the BE2 line to Mg residual doping [5]. Indeed, the MBE reactor has been used to grow GaN light-emitting diodes on the one hand [7] and Mg is used for the ELOG growth on the other hand [4]. A couple of typical GaN excitonic photoluminescence spectra are presented in figure 3.

When the external voltage was applied to the structure, the quenching of excitonic photoluminescence was observed. The character of photoluminescence behaviour with electric field depends on the binding energy of corresponding exciton state as shown in figure 4, where the dependence of the line intensities versus the magnitude of the electric field is plotted.

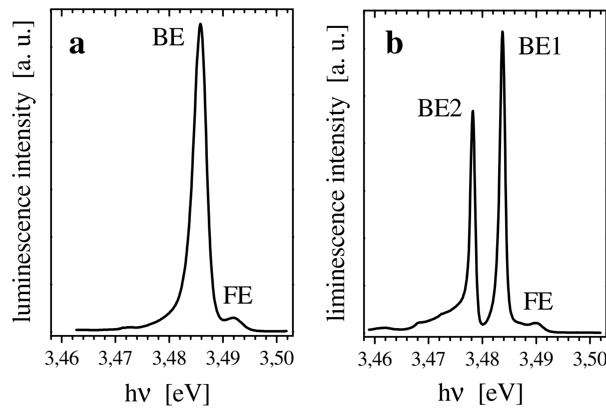

Figure 3. Excitonic luminescence spectra of two different GaN samples. $T = 2$ K.

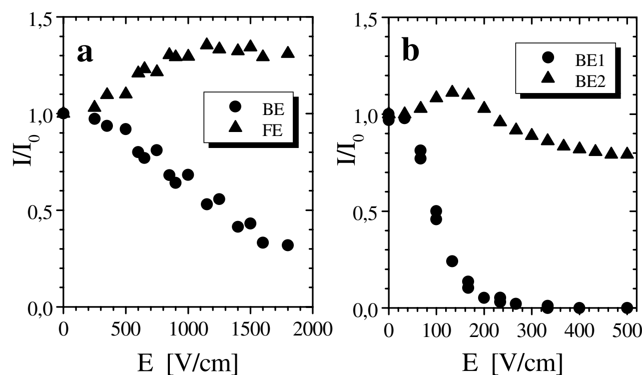

Figure 4. Dependences of the relative intensities of excitonic lines on the applied electric field. (a) MOVPE sample with only one bound exciton line with binding energy $\varepsilon_b = 6.5$ meV; (b) MBE sample with two bound exciton lines with binding energies $\varepsilon_b = 4.5$ meV and $\varepsilon_b = 12$ meV.

One can see that the line with the smallest binding energy (BE for case (a) and BE1 for case (b)) undergoes the most significant quenching. At the same time, in the range of relatively weak electric field, an enhancement of the line with the largest binding energy is observed. When the electric field becomes stronger it is replaced by a saturation (for the FE line) or by a quenching (for the BE2 line).

The observed effect can be explained in terms of the impact ionization model. Hot electrons with the kinetic energy of about the binding energy of the exciton to the impurity centre (or about the exciton binding energy in the case of a free exciton) destroy the excitonic state leading to the quenching of the corresponding emission line. At relatively weak electric field, this effect is only effective for the states with small binding energy. The dissociation of the complex with the lowest binding energy increases the concentration of free excitons that can be trapped on the deeper centres or can contribute to enhance the free exciton line. In a stronger electric field the electrons can acquire the energy required to destroy the complexes with large binding energies, so the intensity of the corresponding line decreases. Practically, in the range of electric field we studied, we only observed the quenching for bound exciton lines with different binding energies (from 4.5 meV to 12 meV). For free excitons we only observed the

saturation of the line intensity. The quenching of such a strongly bound state (binding energy is about 28 meV) needs a much stronger electric field.

A similar effect can be observed in GaN/AlGaN quantum wells. The photoluminescence spectra of QW-structures consist of an intense line of localized exciton and of its rather weak phonon replica. The exciton localization energy can be estimated by a Stokes shift of the luminescence line and appears to be about 20 meV [8]. When the electric field is applied to the sample, the photoluminescence significantly quenches (see figure 5). This effect can also be accounted for by delocalization of excitons under impacts of hot electrons.

4. Data analysis

The equation for the number of excitons N in a system is written:

$$\frac{\mathrm{d}N}{\mathrm{d}t} = -\frac{N}{\tau} + G \tag{1}$$

where G is the generation rate. The probability of exciton (or exciton–impurity complex) decay $(1/\tau)$ in the presence of electric field E is determined by the probability of impact ionization $W(E)$ and by the decay probability due to other radiative and nonradiative processes $1/\tau_0$:

$$\frac{1}{\tau} = \frac{1}{\tau_0} + W(E). \tag{2}$$

Taking into account that the luminescence intensity $I(E) \sim N(E)$, we obtain for the stationary conditions regime the following expression:

$$I(E) = I(0)[1 + W(E)\tau_0]^{-1}. \tag{3}$$

The impact ionization probability $W(E)$ is defined as the integral, over the energies exceeding the ionization energy, of the product of the ionization probability per electron by the density of states, and by the electron energy distribution function $f(\varepsilon)$. Only the latter decreases exponentially with the energy, the first two being power-law functions. The integral is determined by the small vicinity around the ionization threshold and is revealed to be exponentially small. The argument of the exponential determines the electric field dependence of the ionization coefficient. We shall not be interested in the pre-exponential factors, which is allowable when the modulus of the exponent is large. The task is to calculate the small number of electrons having the energy sufficient for ionization and thus determining the ionization probability.

The electron momentum distribution function $f_{\boldsymbol{p}}$ obeys the kinetic equation

$$e\boldsymbol{E}\frac{\partial f_{\boldsymbol{p}}}{\partial \boldsymbol{p}} + \frac{1}{\tau_{im}(\varepsilon_p)}(f_{\boldsymbol{p}} - f(\varepsilon_p)) + \frac{2\pi}{\hbar}\sum_{\boldsymbol{q}} |C_q|^2[(f_{\boldsymbol{p}}(N_q + 1) - f_{\boldsymbol{p}-\hbar\boldsymbol{q}}N_q)\delta(\varepsilon_{\boldsymbol{p}} - \varepsilon_{\boldsymbol{p}-\hbar\boldsymbol{q}} - \hbar\omega_q)$$
$$+(f_{\boldsymbol{p}}N_q - f_{\boldsymbol{p}+\hbar\boldsymbol{q}}(N_q + 1))\delta(\varepsilon_{\boldsymbol{p}} - \varepsilon_{\boldsymbol{p}+\hbar\boldsymbol{q}} + \hbar\omega_q)] = 0 \tag{4}$$

where $\boldsymbol{p}$ is the electron momentum, $\varepsilon_p = p^2/2m$ its energy, m its effective mass; $\boldsymbol{q}$ is the phonon wavevector, ω_q its frequency: $\omega_q = sq$, s is the velocity of sound; $N_q = \exp(\hbar\omega_q/kT - 1)^{-1}$ is the Planck distribution function for phonons; $\tau_{im}(\varepsilon_p)$ is the relaxation time for elastic scattering on impurities and lattice defects; $C_{\boldsymbol{q}}$ is the electron–phonon interaction constant. Only interactions with acoustic or piezoacoustic phonons are taken into account, since the limiting frequency of optical phonons in GaN is larger than the exciton ionization energies. From the conservation laws in the electron–phonon interaction process it follows that only sufficiently large phonon energies are essential and at low temperatures the number of phonons is exponentially small $N_q \approx \exp(-\hbar\omega_q/kT)$. Therefore it can be neglected in comparison with unity. The electron relaxation due to the process of exciton ionization is not taken into

account in (4), being negligible as compared with the relaxation rate caused by interaction with phonons and impurities.

Let us transfer the terms with a minus sign to the right-hand side and then, treating them as an inhomogeneity, rewrite (4) in the form of an integral equation

$$f_p = \int_0^\infty \mathrm{d}t \Bigg\{ \frac{1}{\tau_{im}(\varepsilon_{p-eEt})} f(\varepsilon_{p-eEt}) + \frac{2\pi}{\hbar} \sum_q |C_q|^2 [f_{p-eEt+\hbar q} \delta(\varepsilon_{p-eEt} - \varepsilon_{p-eEt+\hbar q} + \hbar\omega_q) + f_{p-eEt-\hbar q} N_q \delta(\varepsilon_{p-eEt} - \varepsilon_{p-eEt+\hbar q} + \hbar\omega_q)] \times \exp\left[- \int_0^t \mathrm{d}t' \left(\frac{1}{\tau_{im}(\varepsilon_{p-eEt})} + \frac{1}{\tau_{ph}(\varepsilon_{p-eEt})} \right) \right]. \tag{5}$$

The electron relaxation time due to phonons is

$$\tau_{ph}^{-1}(\varepsilon_p) = \frac{2\pi}{\hbar} \sum_q |C_q|^2 \delta(\varepsilon_p - \varepsilon_{p-\hbar q}) \tag{6}$$

and the electron mean free path is given by the expressions

$$I_{ph,im}(\varepsilon_p) = \sqrt{\frac{2\varepsilon_p}{m}} \tau_{ph,im}(\varepsilon_p) \qquad \frac{1}{I(\varepsilon)} = \frac{1}{I_{ph}(\varepsilon)} + \frac{1}{I_{im}(\varepsilon)}. \tag{7}$$

We assume that the electric field is sufficiently strong:

$$\varepsilon_i \gg eEI(\varepsilon_i) \gg kT. \tag{8}$$

As shown by expression (5), the shape of the energy distribution at high energies is determined by the energy distribution at mean (thermal) energies. We assume that at these energies effective energy and momentum relaxation mechanisms are active, so that the distribution function is an equilibrium. The saddle-point-method integration of (5) yields

$$f_p = A_p \exp\left[- \frac{\varepsilon_p}{T} \sin^2\vartheta - \cos\vartheta \int_0^{\varepsilon_p} \frac{\mathrm{d}\varepsilon'}{eEI(\varepsilon')} \right] \tag{9}$$

where A_p is a weaker-than-exponential function of the momentum, ϑ is the angle between the momentum and the electric field. Function (9) is needle shaped; it is elongated in the electric field direction and decreases rapidly in other directions. Averaging of this function over the angles gives the Townsend–Shockley ionization law

$$f(\varepsilon) = \exp\left(- \int_0^\varepsilon \frac{\mathrm{d}\varepsilon'}{eEI(\varepsilon')} \right). \tag{10}$$

The special consideration is necessary in the case of predominance of elastic electron scattering $\tau_{im}(\varepsilon_p) \ll \tau_{ph}(\varepsilon_p)$. It was shown [9] that for different values of electric field different dependences take place. When conditions $kT \ll eE\sqrt{I_{ph}(\varepsilon_i)I_{im}(\varepsilon_i)} \ll s\sqrt{2m\varepsilon_i}$ are met the Townsend–Shockley dependence is manifested

$$f(\varepsilon) = A \exp\left(\int_0^\varepsilon \frac{\sqrt{3}\,\mathrm{d}\varepsilon'}{eE\sqrt{I_{ph}(\varepsilon')I_{im}(\varepsilon')}} \right). \tag{11}$$

Under conditions $kT \ll s\sqrt{2m\varepsilon_i} \ll eE\sqrt{I_{ph}(\varepsilon_i)I_{im}(\varepsilon_i)}$ the distribution function is given by solving the problem of 'hot' electrons, which leads to Davydov–Wolf ionization law

$$f(\varepsilon) = A \exp\left(- \int_0^\varepsilon \frac{6\,\mathrm{d}\varepsilon'}{e^2E^2|(\varepsilon')|_\varepsilon} (\varepsilon') \right) \tag{12}$$

$$I_\varepsilon(\varepsilon_p) = \frac{\sqrt{2m}}{\varepsilon_p^{3/2}} \pi \sum_q |C_q|^2 \omega_q \delta(\varepsilon_p - \varepsilon_{p-\hbar q}). \tag{13}$$

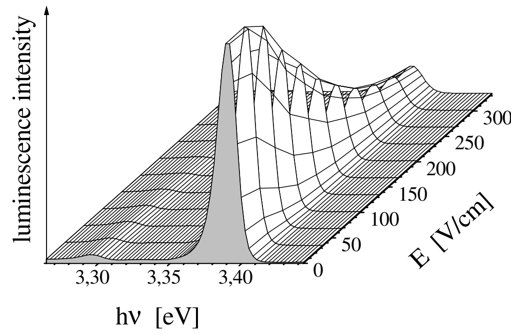

Figure 5. Photoluminescence spectra of GaN/AlGaN quantum well versus applied electric field. The quantum well width is 17 monolayers.

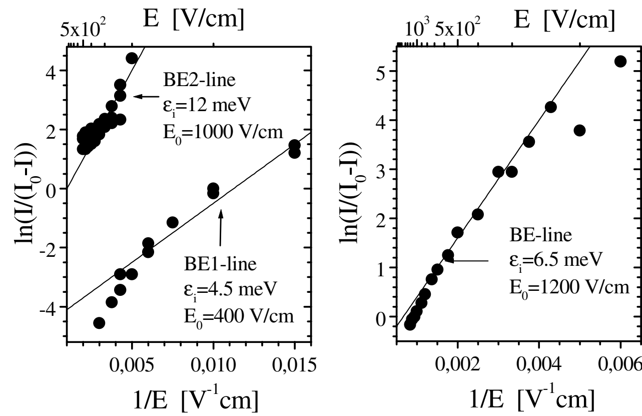

Figure 6. Dependences of the bound exciton luminescence line intensity on the electric field for two different GaN samples. Experimental points are plotted as dependences $\ln[I/(I_0 - I)]$ on E^{-1} to compare with theory. Solid lines are linear fits.

When the electric field increases one could expect the transition from dependence (11) to dependence (12).

Let us analyse the experimental data using the Townsend–Shockley dependence for $W(E) = W_0 \exp(-E_0/E)$, and let us show what information can be obtained from this analysis. For this purpose, the dependence of GaN photoluminescence intensity on the applied electric field is plotted in coordinates $\ln[I/(I_0 - I)]$ versus E^{-1} (see figure 6). The linear fit to experimental points shown in figure 6 by a solid line gives the value of parameter E_0. The values of E_0 obtained by using this procedure for the GaN samples we studied are presented in table 1.

In pure crystals at low temperatures the dominant mechanism of electron relaxation should be the scattering on piezoacoustic phonons. The estimations made using the formulae (6), (7) and (10) and the GaN parameters from [10] give for $\varepsilon_i = 6$ meV the value $E_0 = 160$ V cm^{-1}. This is much less than the value $E_0 = 600$ V cm^{-1} obtained in our experiment. Moreover, in the case of scattering on piezoacoustic phonons, the parameters E_0 should be proportional to the square root of the ionization energy. The data obtained for the MBE sample show that $E_0 \sim \varepsilon_i$. Both these facts indicate that electron relaxation is not only due to scattering on piezoacoustic phonons, but that scattering of electrons on the impurity centres play an

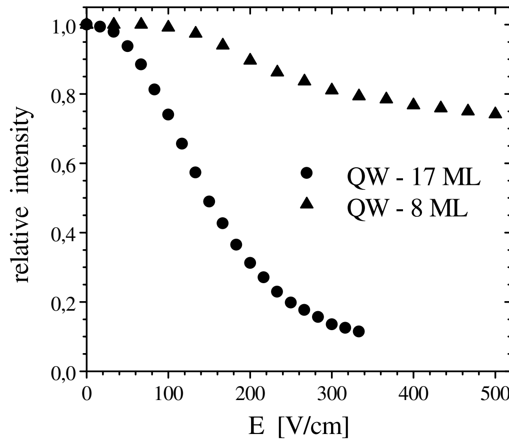

Figure 7. Dependences of luminescence intensity on applied electric field for GaN/AlGaN quantum well samples with different quantum well widths.

essential role in our samples. According to the expression (11), the effective mean free path of hot electrons can be estimated in this case by the formula

$$l_{eff} = \sqrt{3}\varepsilon_i/eE_0. \tag{14}$$

The results of these estimations are presented in the last column of table 1. First we note that ELOG and MBE samples give similar values. Second, we compare these values with those obtained for CdS [11]. In case of pure CdS, the corresponding value is 10^{-4} cm when $\varepsilon_i = 4$ meV. For lower quality CdS, still with $\varepsilon_i = 4$ meV, the corresponding value decreases to 5×10^{-6} cm. This work indicates that the value in today's GaN (2×10^{-5} cm) is not so bad.

Let us now turn to analysis of the data obtained on GaN/AlGaN quantum well structures. The dependences of luminescence intensity on an electric field for two GaN/AlGaN samples with quantum well width of 17 monolayers and 8 monolayers are presented in figure 7. One can see that the quenching effect is much stronger in the sample with a wider well. The main reason for this phenomenon is that the exciton lifetime decreases significantly with decreasing of the well width [12]. According to (3), the decreasing of τ_0 leads to the decreasing of the influence of $W(E)$ on the luminescence intensity.

The linear fit to the dependences $\ln[I/(I_0 - I)]$ versus E^{-1} give the values of parameter E_0: $E_0 = 350$ V cm^{-1} and $E_0 = 480$ V cm^{-1} for 17 ML and 8 ML QWs respectively. Taking the value of exciton localization energy $\varepsilon_l = 20$ MeV, we obtain the following values of the mean free path of hot electrons: $l_{eff} = 1 \times 10^{-4}$ cm for 17 ML QW and $l_{eff} = 0.7 \times 10^{-4}$ cm for 8 ML one. These values are significantly larger than that of bulk GaN. When calculating the mean free path of electrons in quantum wells one should take

Table 1. The values of parameter E_0 and of the mean free path of hot electrons l_{eff} for different GaN samples.

Sample, line	ε_i (meV)	E_0 (V cm^{-1})	l_{eff} (cm)
ELOG, BE1 line	6.5	600	2×10^{-5}
MBE, BE1 line	4.5	400	2×10^{-5}
MBE, BE2 line	12	1000	2×10^{-5}
MOVPE, BE line	6.0	1500	7×10^{-6}

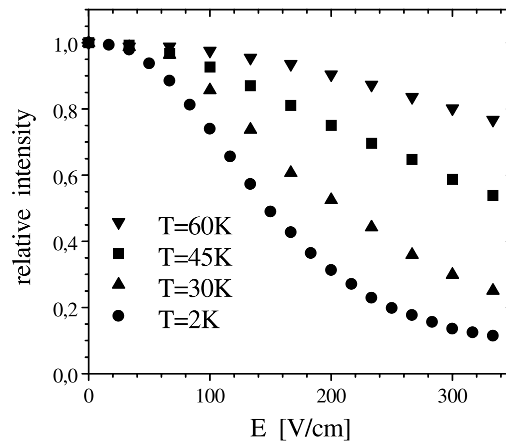

Figure 8. Dependences of luminescence line intensity on applied electric field for different temperatures. GaN/AlGaN quantum well width is 17 monolayers.

into account that, in the scattering process, the change of electron momentum in a direction perpendicular to the well plane cannot be more than $\hbar/d$, where d is the well thickness. Therefore in the two-dimensional case the scattering probability of the electron decreases and the mean free path increases by a factor of $\sqrt{2m\varepsilon_i}d/\hbar$. The presence of this factor explains the observed relation between mean free paths of hot electrons in GaN films and GaN/AlGaN QWs.

The temperature dependence of the impact ionization effect in QWs was studied in the temperature interval from 2 K to 60 K. When the temperature increases the quenching effect becomes weaker (see figure 8). Similarly to the experiment on the samples with different well width, this can be explained by the decreasing of exciton lifetime with increasing of temperature. At the same time, the analysis of experimental data using the Townsend–Shockley dependence for $W(E)$ have shown that there is no temperature dependence of parameter E_0 in agreement with the theory, so the whole decrease in the impact ionization effect is accounted for by the variation of τ_0 with temperature.

5. Conclusion

In conclusion, we wish to emphasize the fact that in non-intentionally doped GaN epilayers grown by MOVPE, ELOG and MBE, the electron energy and momentum relaxation is ruled by the scattering on impurities, as an evidence of a significant departure from the pure crystal situation where the dominant mechanisms is scattering by acoustic phonons. In GaN/AlGaN quantum wells the mean free paths of hot electrons appear to be an order of magnitude larger than that of GaN films due to a decrease in the scattering probability of an electron in the two dimensional case.

Acknowledgments

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