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Dopant-Induced Electric Fields and Their Influence on the Band-Edge Absorption of GaN

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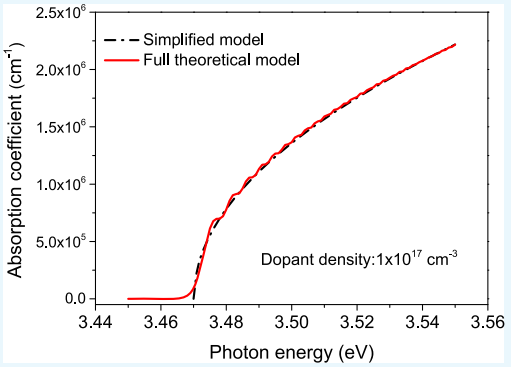
ABSTRACT: Dopant-induced local electric fields and their influence on the band-edge absorption of GaN are theoretically examined. For dopant-induced electric field distribution, it is derived with Bayes' rule. For the average electric field strength, it is revealed to be quite strong, i.e., in an order of 10^4 V/cm in GaN with a fairly low dopant density. On the basis of the Franz−Keldysh mechanism, influence of the dopant-induced electric fields on the band-edge absorption coefficient of GaN is then investigated. Without any adjustable parameters, absorption coefficients of GaN are computed and in good agreement with the available experimental values.



I. INTRODUCTION

Gallium nitride (GaN) is a wide band gap semiconductor that can be used in the production of new power electronics, RF components, and light-emitting devices. As the Si technology is approaching its ultimate limits, GaN technology is on the rise for today and tomorrow. To further prompt the GaN technology demands understanding and manipulating GaN in new ways, such as doping control at the atomic level and the related effects. For instance, in a recent study, Tuomisto et al. have proved a big promise of beryllium doping for GaN power electronics if the Be dopants in GaN and their electronic properties can be fully controlled.[1] Actually, impurity doping plays a vital role in the fabrication of almost all the semiconductor devices,[2] not only in the GaN-based devices.[3−5] Moreover, further development and optimization of GaN-based electronic and optoelectronic devices require control over the doping of GaN-based materials. To fully control the doping of GaN, a deeper fundamental understanding of the dopant issues is necessary.[6] For example, there have been very few studies on electric fields induced by ionized dopants and relevant effects in GaN so far. In particular, the present understanding of the relationship between dopant-induced electric fields and band-edge absorption coefficients in GaN is very limited. In the very early studies, Redfield theoretically investigated the distribution of electric fields in solid-containing dopants and the influence on the optical absorption edge of the solid.[7] His major conclusions are that in nonmetallic solids, the dopant-induced local electric fields are substantial and their effects are significant. It is obvious that taking an explicit consideration to the dopant-induced local electric fields and their effects in GaN is of scientific and technical significance and is hence highly desirable. Furthermore, we may obtain useful information of dopant density in GaN by establishing the relationship between the band-edge absorption coefficients and dopant density.

In this article, we theoretically investigate the strength and distribution of electric fields in GaN with different densities of dopants. Then, we compute the influence of dopant-induced fields on the absorption coefficients of GaN in terms of the Franz−Keldysh effect.

II. THEORY

II.I. Dopant-Induced Electrical Fields and Distribution. To evaluate the electric fields of dopants in a crystal of static dielectric constant ε_r, we assume that N singly charged dopants (point defects) per m^{33} are randomly distributed within the crystal. According to the Coulomb law, a local electric field by a singly charged dopant may be represented as

$$F_0 = \frac{1}{4\pi\varepsilon_r\varepsilon_0}\frac{|e|}{r_0^2} \tag{1}$$

where e is the electron charge, ε_0 is the vacuum dielectric constant, and r_0 is the radius of a sphere whose volume is the mean volume of a dopant. Thus, r_0 may be defined by

$$\frac{4}{3}\pi r_0^3 = \frac{1}{N} \text{ or } r_0 = 0.62N^{-1/3} \tag{2}$$

For many purposes, as stated by Redfield,[7] it is the most useful way to define a probability $W(F)dF$ of finding a field of magnitude F. When normalized to unity, it is the fraction of the volume of a crystal occupied by fields in the range of F to $F + dF$. Under a first approximation, only the Coulomb field of

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the nearest charge ion is taken into account for the calculation of electric field distributions. This "nearest ion distribution" can be readily found from the position distribution function of random points.[8] Herein, we use a more general approach (i.e., the conditional probability theorem) to derive the expression of $W(F)\mathrm{d}F$. Selecting a dopant (radiating atom) as the center, we describe a sphere of radius r about it. $P(r)$ denotes the probability of finding at least one ion (charged dopant) within it. To obtain $P(r)$, we may use the conditional probability theorem, also widely known as Bayes' rule. By Bayes' rule, we have

$$P(r|r + \mathrm{d}r) = \frac{P(r + \mathrm{d}r|r)P(r)}{P(r + \mathrm{d}r)} \tag{3}$$

where $P(r|r + \mathrm{d}r)$ is a conditional probability: the likelihood of an event r to occur given that event $r + \mathrm{d}r$ is true. Similarly, $P(r + \mathrm{d}r|r)$ is also a conditional probability: the likelihood of an event $r + \mathrm{d}r$ to occur given that event r is true.

According to the definition of $P(r)$, we can clearly conclude that $P(r + \mathrm{d}r|r) = 1$, so that eq 3 can be reduced as

$$P(r|r + \mathrm{d}r) = \frac{P(r)}{P(r + \mathrm{d}r)} = \frac{P(r)}{P(r) + P'(r)\mathrm{d}r} \tag{4}$$

where $P'(r)$ is the first-order derivative of $P(r)$.

Considering that dopants are randomly distributed in the crystal lattice, we also have

$$P(r|r + \mathrm{d}r) = \frac{P(r)}{P(r) + P(r^{\mathrm{C}})P(\mathrm{d}r|_r)} = \frac{P(r)}{P(r) + (1-P(r))4\pi r^2 N\mathrm{d}r} \tag{5}$$

where r^{C} is the event of complement of r, $P(\mathrm{d}r|_r)$ is the probability of an event that there is at least one dopant within the spherical shell of radius r and thickness $\mathrm{d}r$.

Combining eqs 4 and 5, we can get

$$P'(r)\mathrm{d}r = (1 - P(r))4\pi r^2 N\mathrm{d}r \tag{6}$$

Considering the boundary conditions $P(0) = 0$ and $\int_0^\infty \mathrm{d}P(r) = 1$, we can yield

$$P(r) = 1 - \mathrm{e}^{-4\pi/3Nr^3}, \text{ and } \mathrm{d}P(r) = 4\pi Nr^2\mathrm{e}^{-4\pi/3Nr^3}\mathrm{d}r \tag{7}$$

In terms of r_0 and F_0 described by eqs 2 and 1, respectively, we can rewrite eq 7 as

$$\mathrm{d}P(r) = \mathrm{e}^{-(r/r_0)^3}\mathrm{d}\left(\frac{r}{r_0}\right)^3 \tag{8}$$

$$\mathrm{d}P(F) = \frac{3}{2F}\left(\frac{F_0}{F}\right)^{3/2}\mathrm{e}^{-(F_0/F)^{3/2}}\mathrm{d}F \tag{9}$$

Actually, eq 9 is just the same as the nearest ion distribution in weakly ionized plasmas.[8]

By letting $\beta = F/F_0$, eq 9 can be rewritten as

$$\mathrm{d}P(F) = \frac{3}{2}\beta^{-5/2}\mathrm{e}^{-\beta^{-3/2}}\mathrm{d}\beta = W(\beta)\mathrm{d}\beta \tag{10}$$

and

$$W(\beta) = \frac{3}{2}\beta^{-5/2}\exp[-\beta^{-3/2}] \tag{11}$$

II.II. Band-Edge Absorption in the Presence of Dopant-Induced Electric Fields. To evaluate the effects of the electric fields of charged dopants on the band-edge absorption coefficients of the semiconductors, Redfield[7] theoretically treated it by invoking the analogy with the Franz–Keldysh effect.[9,10] Redfield's theoretical treatment requires making the following approximations:[7]

(i) All transitions are assumed to allow band-to-band transitions. The excitonic effect is totally ignored.

(ii) The electric field is treated as a uniform field in every small but macroscopic volume element of the crystal.

Under these assumptions, the total absorption coefficient may be computed by

$$\alpha(\omega) = \int_0^\infty A(\omega, F)W(F)\mathrm{d}F \tag{12}$$

where $A(\omega,F)$ is the expression describing the local absorption coefficient for angular frequency ω in the presence of a field F, whereas $W(F)$ is the probability of finding an electric field of magnitude F, which is found in eq 11.

For $F \neq 0$, $A(\omega,F)$ may be expressed by[9]

$$A(\omega, F) = \sqrt{\omega_{\mathrm{F}}}\sum_{n=0}^{\infty}\frac{\Gamma(3/2)}{3n!\Gamma(7/6 - n/3)}\left(\frac{\omega - \omega_{\mathrm{g}}}{\omega_{\mathrm{F}}}\right)^n \tag{13}$$

Here, ω_{F} is defined by

$$\omega_{\mathrm{F}} = \left(\frac{e^2F^2}{12\hbar\mu}\right)^{1/3} \tag{14}$$

$\mu = \frac{m_{\mathrm{e}}{}^* m_{\mathrm{h}}{}^*}{m_{\mathrm{e}}{}^* + m_{\mathrm{h}}{}^*}$ is the reduced mass with $m_{\mathrm{e}}{}^*$ and $m_{\mathrm{h}}{}^*$ as the effective masses of the electrons and holes in the conduction and valence bands, respectively. $\hbar\omega_{\mathrm{g}} = E_{\mathrm{g}}$ gives the fundamental band gap of the crystal, whereas $\Gamma(z) = \int_0^\infty x^{z-1}\mathrm{e}^{-x}\mathrm{d}x$ is the γ function.[11] For $z = 3/2$, $\Gamma(3/2) = \sqrt{\pi}/2$.

At $\omega = \omega_{\mathrm{g}}$, eq 13 may become simply

$$A(\omega_{\mathrm{g}}, F) = \sqrt{\omega_{\mathrm{F}}}\frac{\Gamma(3/2)}{3\Gamma(7/6)} = \frac{1}{6\Gamma(7/6)}\sqrt{\pi\omega_{\mathrm{F}}} \tag{15}$$

For the absorption below the band gap, the computation becomes considerably more difficult and has to be performed numerically. Furthermore, the infinite range of fields in the integration of eq 12 precludes the use of any of the approximate formulae. In fact, to obtain a rigorous expression for $A(\omega,F)$, Tharmalingam reformulated Franz's result and yielded[12]

$$A(\omega, F) = (\omega_{\mathrm{F}})^{1/2}\int_{(\omega_{\mathrm{g}}-\omega)/\omega_{\mathrm{F}}}^{\infty}|\mathrm{Ai}(z)|^2\mathrm{d}z \tag{16}$$

where $\mathrm{Ai}(z) = \frac{1}{\sqrt{\pi}}\int_0^\infty \cos\left(uz + \frac{1}{3}u^3\right)\mathrm{d}u$ is the Airy function.[13] Since the Airy function satisfies the differential equation

$$\frac{\mathrm{d}^2\mathrm{Ai}(z)}{\mathrm{d}z^2} = z\mathrm{Ai}(z) \tag{17}$$

The last integral in eq 16 can be evaluated to give

$$\int_\theta^\infty |\mathrm{Ai}(z)|^2\mathrm{d}z = -\theta|\mathrm{Ai}(\theta)|^2 + |\mathrm{Ai}'(\theta)|^2 \tag{18}$$

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where prime denotes derivative of the Airy function with respect to the argument.

It should be noted that to convert $A(\omega,F)$ to the absolute absorption coefficient $\alpha(\omega,F)$, all that is needed is to multiply it by an appropriate factor R. It may be represented by[12]

$$R(\omega) = \frac{2e^2C_0^2}{\hbar\omega cnm_0^2}\left(\frac{2\mu}{\hbar}\right)^{3/2} \tag{19}$$

where C_0 involves the matrix element having the dimensions of momentum, c is the speed of light in vacuum, n is the refractive index of the crystal, and m_0 is the rest mass of free electrons.

III. RESULTS AND DISCUSSION

Figure 1 shows the average electric fields F_0 (bottom) induced by dopants and the mean radii r_0 (top) of one dopant in GaN,

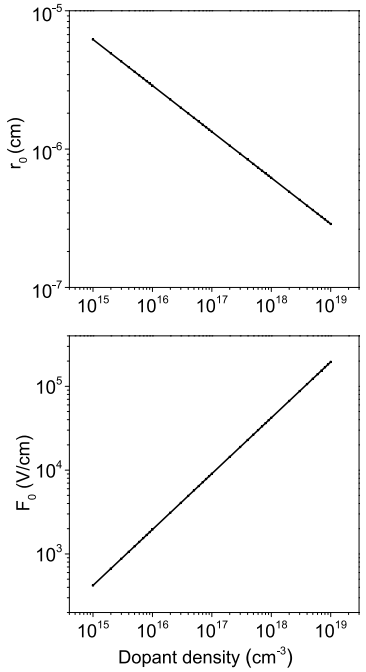

Figure 1. Calculated mean radii of a dopant (top), and average electric fields vs dopant density.

calculated by eqs 1 and 2, respectively. As expected, the average electric field induced by ionized dopants monotonically increases with the increasing dopant density, whereas the mean radius of a dopant decreases in a monotonic manner. The dopant-induced local electric field in a GaN of fair purity is substantially strong, e.g., $F_0 \sim 10^4$ V/cm for $N = 10^{17}$ cm^{-3} dopants. At this dopant density, the mean radius of one dopant is about 20 nm, which means that the local field is distributed over such a range. Clearly, the dopant-induced electric fields in GaN cannot be ignored, and they may have substantial impact on the photophysical properties of GaN as proved later. Parameters of GaN adopted in the calculations are tabulated in Table 1.

Figure 2 shows the distribution of dopant electric fields described by eq 11. From the definition of $\beta = F/F_0$, it can be well-justified that the magnitudes of the electric fields are distributed most probably around F_0, which acts like a scale factor.[7] For small and large values of β, respectively, eq 11 can have the two alternate expressions

Table 1. Parameters of GaN Used in the Calculations

electron effective mass	$0.20m_0$[14]
hole effective mass	$0.8m_0$[15]
band-edge absorption coefficient	8×10^6 m^{-1} [16]
dielectric constant (static)	8.9[17]
refractive index	2.6822[18]
energy band gap	3.47 eV[14]
electron rest mass m_0	9.109×10^{-31} kg
electron charge	1.602×10^{-19} C
reduced Planck constant	1.05457×10^{-34} J s

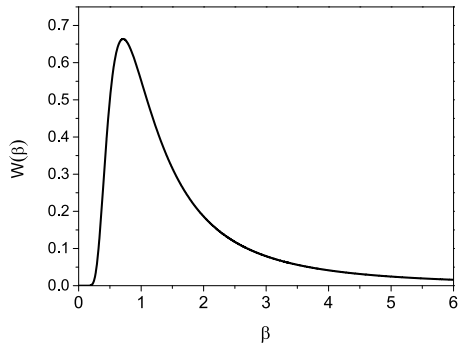

Figure 2. Probability distribution of the dopant electric fields vs $\beta = F/F_0$.

$$W(\beta) = \begin{cases} (4/3\pi)\beta^2(1 - 0.463\beta^2 + 0.1227\beta^4\cdots) \\ 1.496\beta^{-5/2}(1 + 5.107\beta^{-3/2} + 14.93\beta^{-3} + \cdots) \end{cases} \tag{20}$$

In particular, the probability distribution of high electric fields decreases as $\beta^{-5/2}$.

From transmission measurements, Muth et al. obtained the band-edge absorption coefficient of GaN, as listed in Table 1.[16] We calculated the band-edge absorption coefficients of GaN under the action of different electric fields, as shown in Figure 3. Calculated band-edge absorption coefficient of GaN at an

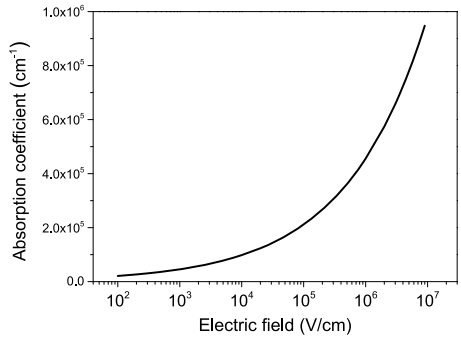

Figure 3. Calculated band-edge absorption coefficients of GaN vs electric field. Note that the curve is plotted in semilogarithmic scale.

electric field of 6×10^3 V/cm is comparable to their measured value. As shown later, this field corresponds to a dopant density of 4×10^{16} cm^{-3}. Actually, the dopant concentration in GaN is fairly low. It is known that nominally undoped GaN shows a usually high n-type conductivity with an electron concentration of 4×10^{16} to 9×10^{18} cm^{-3}.[19] Furthermore, the theoretical absorption coefficient of GaN rapidly increases with

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increasing electric field, which can be understood as the electric-field-induced Stark effect.

Figure 4 illustrates the calculated band-edge absorption coefficients of GaN vs dopant density. It can be seen that the

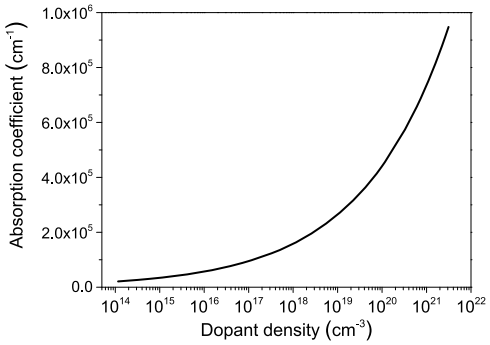

Figure 4. Calculated band-edge absorption coefficients of GaN vs dopant density. Note that the curve is plotted in a semilogarithmic scale.

band-edge absorption coefficient of GaN shows a strong dependence on dopant density. In fact, the strong dependence of the band-edge absorption coefficient of GaN on dopant density also reflects the significant effect of electric fields induced by ionized dopants.

It is instructive to discuss the several limiting cases for absorption:

(i) For $\omega > \omega_g$ and near the band edge

$$\alpha \cong R(\omega)(\omega_F)^{1/2}\left[\left(\frac{\omega-\omega_g}{\omega_F}\right)^{1/2} + \int_0^\infty |\mathrm{Ai}(z)|^2 dz\right] \rightarrow R(\omega)(\omega-\omega_g)^{1/2} \text{ as } F \rightarrow 0 \quad (21)$$

Obviously, for $F \rightarrow 0$, and $\omega > \omega_g$, we obtain the familiar classic expression for absorption coefficient with the square root dependence on the photon energy or frequency,[20] of course, relative to the band gap.

(ii) For $\omega < \omega_g$ and $\frac{\omega_g-\omega}{\omega_F} \gg 1$, using asymptotic series of the Airy function, $\mathrm{Ai}(z) = \frac{z^{-1/4}}{2\sqrt{\pi}}e^{-2/3z^{3/2}}\left(1-\frac{5}{48}z^{-3/2}\right)$ and $\mathrm{Ai}'(z) = -\frac{z^{1/4}}{2\sqrt{\pi}}e^{-2/3z^{3/2}}\left(1+\frac{63}{432}z^{-3/2}\right)$

we can obtain

$$\alpha \approx R(\omega)\frac{{\omega_F}^{3/2}}{8\pi(\omega_g-\omega)}\exp\left[-\frac{4}{3}\left(\frac{\omega_g-\omega}{\omega_F}\right)^{3/2}\right] \quad (22)$$

hence giving an exponential decaying tail in the long wavelength region below the band gap. Such an exponential tail in the absorption coefficient for $\omega \ll \omega_g$ is consistent with the well-known Urbach's tail rule.[21]

Figure 5 shows the theoretical absorption curves of GaN with full theoretical model (i.e., eqs 12 and 16) and a simplified model (i.e., eq 21) for a relatively fair low density of dopant, respectively. For $\omega \gg \omega_g$, i.e., well above the band gap, the two curves coincide with each other. For $\omega > \omega_g$, good agreement between the full theoretical model and simplified formula is still seen. But some oscillatory structures can be observed on the absorption curve by the full theoretical model

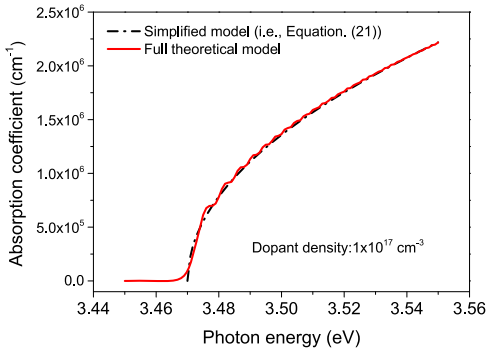

Figure 5. Calculated absorption coefficients of GaN vs photon energy with a full theoretical model and a simplified model (i.e., eq 21).

caused by the oscillatory nature of the Airy function. For $\omega < \omega_g$, i.e., below the band gap, however, there is a substantial difference between the two theoretical curves. The curve by the simplified model directly goes down to zero at the band edge ($\omega = \omega_g$), whereas the curve predicted by the full theoretical model roughly exhibits an exponential decaying tendency. Obviously, the simplified model is only valid for above the band gap and fails to predict the absorption curves at the band gap and below band gap. To have a better and clearer inspection into the exponential decaying tendency below band gap, we enlarge the absorption curve (solid line) below band gap with the full theoretical model in Figure 6. Meanwhile, we

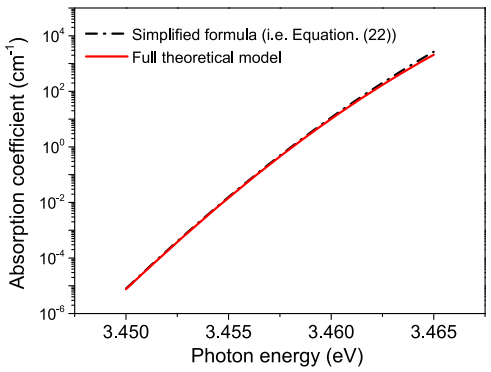

Figure 6. Enlarged absorption curves of GaN below band gap with the full theoretical model and a simplified formula (i.e., eq 22). The absorption coefficients are depicted in logarithmic scale such that exponential decaying tendency can be better seen.

also plot a curve calculated with a simplified formula described by eq 22 for comparison. Clearly, both full model and simplified formula can predict a nearly identical exponential decaying tendency for absorption coefficients below band gap. The exponential decaying tail is just expected by Urbach's law.[21]

It should be noted that no adjustable parameters are adopted in the above calculations. However, the exciton effect on the absorption coefficient of GaN is not taken into account. In the absence of an electric field, the exciton effect has been considered by Elliot.[22] In the presence of an electric field, theoretical treatment on the absorption coefficient of excitons becomes quite complicated.[23] For GaN considered here, its exciton effect shall be weak at room temperature because the binding energy of excitons in GaN is about 25 meV.[24] At room temperature (i.e., 295 K or the corresponding thermal energy ~25.4 meV), most of the excitons may be thermalized into free

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electrons and holes. It is thus reasonable for one not to consider the exciton effect in the room-temperature absorption of GaN.

Another noticing point is that the polar electric field caused by longitudinal optical phonons in GaN with wurtzite crystal structure is not taken into account. Such a polar field may result in some periodic structures, i.e., phonon sidebands in absorption and luminescence spectrum.

Finally, we present a direct comparison between our theoretical curve (solid circles) and Muth et al.'s experimental data (star symbols) for GaN, as shown in Figure 7. Overall, a

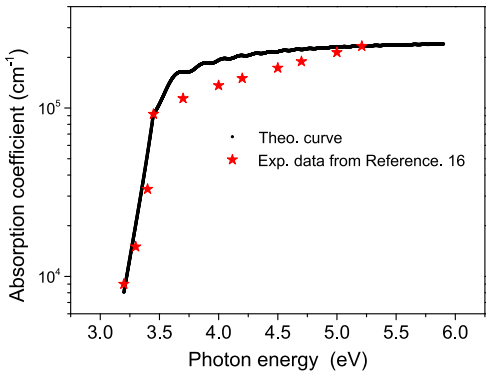

Figure 7. Theoretical absorption spectrum (solid circles) and experimental data (star symbols) by Muth et al. for GaN.[16] Note that the absorption coefficients are shown in logarithmic scale. (Reproduced from [Appl. Phys. Lett. 71, 2572 (1997); https://doi.org/10.1063/1.120191], with the permission of AIP Publishing.)

reasonable agreement between theory and experiment is achieved for a dopant density of 3×10^{20} cm^{-3}. In particular, very good agreement exists for photon energies below the band gap, indicating that the dopant-induced electric fields indeed play a major role in determining the below-band-gap absorption coefficients of GaN. However, above the band gap, a deviation between theory and experiment is observed. This discrepancy could be caused by an overestimation of the absorption coefficients of GaN by the theoretical model.

IV. SUMMARY

We employ the Bayes' rule to derive the probability distribution of the dopant-induced local electric fields in solid, and then apply it to calculate the influence of fields on the absorption coefficients of GaN with different dopant densities. It is shown that a strong electric field of ~10^4 V/cm may exist in an about 20 nm range in GaN with a fairly low dopant density and has a significant impact on the band-edge absorption coefficients of GaN on the basis of the Franz–Keldysh mechanism. It is unveiled that the band-edge absorption coefficient of GaN increases strongly with increasing dopant density. Moreover, analytical expressions of absorption coefficients of GaN in some limiting cases are approximated using the known properties of the Airy function.

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Notes

The authors declare no competing financial interest.

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