Landau levels and magnetopolaron effect in dilute GaAs:N

P. M. Krstajić
Departement Fysica, Universiteit Antwerpen,
Groenenborgerlaan 171, B-2020 Antwerpen, Belgium
Concordia University, 7141 Sherbrooke Ouest, Montréal, Canada.

F. M. Peeters
Departement Fysica, Universiteit Antwerpen,
Groenenborgerlaan 171, B-2020 Antwerpen, Belgium

M. Helm
Institute of Ion Beam Physics and Materials Research,
Forschungszentrum Dresden-Rossendorf,
P.O. Box 510119, 01314 Dresden, Germany

The magnetic-field dependence of the energy spectrum of GaAs doped with nitrogen impurities is investigated. Our theoretical model is based on the phenomenological Band Anticrossing Model (BAC) which we extended in order to include magnetic field and electron-phonon interaction. Due to the highly localized nature of the nitrogen state, we find that the energy levels are very different from those of pure GaAs. The polaron correction results in a lower cyclotron resonance energy as compared to pure GaAs. The magneto-absorption spectrum exhibits series of asymmetric peaks close to the cyclotron energy $\hbar\omega_c$.

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I. INTRODUCTION

Contemporary epitaxial growth techniques provide the possibility for synthesis of high quality semiconductor alloys and/or elemental materials. Recently, there has been growing interest in N doped GaAs, due to its possible applications for long wavelength optoelectronic devices[1]. In GaAs$_{1-x}$N$_x$ substitutional nitrogen has the same valence state as As, thus forming an isoelectronic impurity. Experimental data have unambiguously shown that the effects of nitrogen incorporation are at least three-fold: a) reduction of the fundamental band-gap[2, 3]; b) change in the electron effective mass[4, 5]; and c) decrease in the electron mobility[6].

Substitutional nitrogen in GaAs forms a resonant level, above the conduction minimum[7] ($E_L = 0.23$eV), having A$_1$ (spherical) symmetry. Since N(2s$^2$2p$^3$) has the same valence state as As(4s$^2$4p$^3$) they differ mainly in their local pseudo-potentials resulting in interaction that is predominantly short-range[8, 9]. It is found that the position of this resonant level, $E_L$, does not change for nitrogen concentration up to $x = 3\%$, and the bowing of the conduction minimum follows a simple square root law for small $x$. Therefore one may expect that a phenomenological approach using perturbation theory is sufficient to explain, for instance, the reduction of the band-gap. However, this is true only in part, as the significant difference in bond lengths between Ga-As and Ga-N affects next-nearest neighbours[10], so that the impurity potential has a part which is of intermediate range. What is more, for higher concentration of nitrogen ($x > 1\%$). GaAs:N is classified as a semiconductor alloy, when the use of Virtual Crystal Approximation (VCA) is questionable, and partial collapse of the Brillouin zone is expected. Some authors[12, 13] distinguish three ranges for the molar concentration: 1) ultradilute ($x < 0.01\%$), 2) dilute (0.01% $< x < 1\%$), 3) semiconductor alloy ($x > 1\%$). In this paper we will confine our treatment of GaAs doped with moderately low concentration of N ($x = 0.08\%$) which corresponds to case 2). We will investigate the effect of a high magnetic field and of electron-phonon interaction on the energy spectrum and on the magneto-absorption spectrum of GaAs$_{1-x}$N$_x$ that has not been considered up to now.

The paper is organized as follows. In Sect. II we present our theoretical formalism which is based on the phenomenological Band Anticrossing Model (BAC). It is shown how to derive the energy spectrum in the presence of magnetic field in bulk GaAs:N. In the following section, Sect. III we discuss the influence of the electron-phonon interaction on the fundamental transition energy, i.e. the difference between the first two Landau levels. In Sect. IV theoretical estimates of the absorption spectrum are given for two different values of temperature and magnetic field. In the last section, Sect. V we summarize the results and present our conclusions.

II. THEORETICAL FORMALISM

The Band Anticrossing Model (BAC) will be employed to determine the energy levels in GaAs:N under an applied magnetic field $B$ with inclusion of electron-phonon interaction. Within this model the interaction between the nitrogen state and the conduction states is characterized by a single value $C_N$, to be defined later. This
is possible in dilute GaAs:N where the overlap of neighboring nitrogen wavefunctions is negligible. Then the total wavefunction can be written as a linear combination of extended states $\Psi^{(0)}_{C}$ of pure GaAs, and the impurity wavefunction $\Psi^{(0)}_{L}$,

$$\Psi(r) = \alpha \Psi^{(0)}_{C}(r) + \beta \Psi^{(0)}_{L}(r).$$  \hspace{1cm} (1)

It is convenient to represent the conduction band wavefunction $\Psi^{(0)}_{C}(r)$ in the basis of Wannier wavefunctions, $a_{C}(r - R_{j})$, centered around the sites of the crystal $R_{j}$

$$\Psi^{(0)}_{C}(r) = \frac{1}{\sqrt{M}} \sum_{j} a_{C}(r - R_{j}) e^{i k \cdot R_{j}}, \hspace{1cm} (2)$$

In the above expression $M$ is the number of Ga-As pairs in arbitrary large volume of the crystal. The matrix element between the nitrogen state and conduction states of pure GaAs is

$$\langle \Psi^{(0)}_{L}, \Delta V(r) \rangle \Psi^{(0)}_{C} = \sqrt{x} C_{N}, \hspace{1cm} (3)$$

where $x$ is the concentration of nitrogen impurities in the semiconductor. Eqs. (1)-(3) give rise to a two-level like secular equation,

$$\begin{vmatrix} E - E_{C}(k) \sqrt{x} C_{N} \\ \sqrt{x} C_{N} \end{vmatrix} = 0,$$  \hspace{1cm} (4)

where $E_{C}(k)$ is the energy dispersion of the conduction band, and $E_{L}$ is the N-impurity level. The solutions of Eq. (4) are

$$E_{\pm} = \frac{1}{2} \left\{ E_{C} + E_{L} \pm \sqrt{(E_{C} - E_{L})^{2} + 4 x C_{N}^{2}} \right\}. \hspace{1cm} (5)$$

The value of $C_{N}$ is estimated to be around 2.7eV by fitting to experimental data[3]. The envelope wave function of a nitrogen impurity can be approximated by a Gaussian-like function having only one parameter, the localization radius $a$,

$$\Psi_{L}(r) = \frac{1}{a^{3/2} \pi^{3/4}} e^{- (r - r_{c})^{2} / 2a^{2}}. \hspace{1cm} (6)$$

In the presence of a magnetic field $B$, the conduction band splits in a series of Landau-like levels whose energy spectrum is modified by the presence of the short range impurity potential $\Delta V(r)$,

$$E_{n \pm} = \frac{1}{2} \left\{ E_{Cn} + E_{L} \pm \sqrt{(E_{Cn} - E_{L})^{2} + 4 x C_{N}^{2}} \right\}. \hspace{1cm} (7)$$

The notation $E_{Cn}$ pertains to the pure Landau levels $E_{Cn} = (n + \frac{1}{2}) \hbar \omega_{c} + \hbar^{2} k_{z}^{2} / (2m^{*})$. From now on, index $n$ will refer to pure Landau levels, while $n_{1}$ and $n_{2}$ will refer to lower and upper subband branches ($n-$ and $n+$ in Eq. 4). In the absence of impurities, the conduction (extended) wavefunctions have the form of a linear harmonic oscillator, and for the magnetic field orientation along the $z$ axis and choosing the Landau gauge $A = (-B y, 0, 0)$, their explicit form is as follows

$$\Psi^{LL}_{n}(r) = N_{n} e^{-\frac{(x - y_{0})^{2}}{2\sigma^{2}}} H_{n} \left( \frac{y - y_{0}}{l_{c}} \right) e^{i(k_{z} x + k_{z} z)}. \hspace{1cm} (8)$$

Notation $H_{n}$ corresponds to the $n^{th}$ order Hermitian polynomial, and $y_{0}$ is the $y-$ coordinate of the center of the orbit, while the normalization constant depends on the cyclotron orbit $l_{c}$, and is given by $N_{n} = (\sqrt{\pi} c^{2 n!} n!)^{-1/2}$.

The final wavefunction for the $n^{th}$ lower state has the form:

$$\Psi_{n_{1}}(r) = \alpha_{n_{1}} \Psi^{ LL}_{n_{1}}(r) + \beta_{n_{1}} \Psi_{L}(r). \hspace{1cm} (9)$$

![FIG. 1: Energy levels in GaAs:N with nitrogen concentration of x = 0.08%, as a function of magnetic field. The levels are derived from Landau levels, whose indices are shown near the different curves. The spectrum consists of two branches, one of which is below the nitrogen level, $E_{L}$, while the other one is above. The interaction with optical phonons is neglected for the moment to emphasize the influence of the magnetic field alone. The spectrum splits into two parts with respect to the isolated, localized level $E_{L}$. The lower branch of the levels pin to the value of $E_{L}$ for high values of magnetic field $B$. At this point it would be useful to inspect the behavior of the two branches, $E_{n_{1}}$ and $E_{n_{2}}$. Eq. (7) for small and large values of magnetic field. For small magnetic fields one has the following asymptotic expressions:](image-url)
\[ E_{n_2} = \frac{1}{2} (E_L + D) + A_2 \left( n_2 + \frac{1}{2} \right) \hbar \omega_c + B_2 \left( n_2 + \frac{1}{2} \right)^2 (\hbar \omega_c)^2 + O((\hbar \omega_c)^3), \] (10b)

where \( D = \sqrt{E_L^2 + 4x_C^2} \) and the coefficients \( A_1, A_2 \) and \( B_1, B_2 \) are given by the expressions

\[
A_1 = \frac{1}{2} \left( 1 + \frac{E_L}{D} \right), \quad B_1 = \frac{1}{2} \left( \frac{E_L^2}{D^3} - \frac{1}{D} \right), \quad A_2 = \frac{1}{2} \left( 1 - \frac{E_L}{D} \right), \quad B_2 = -B_1. \tag{11}
\]

The reduction of the bandgap is manifested in Eq. (10a), as the zeroth order term, and is equal to \( (E_L - D)/2 \). The numerical values of \( A_1, A_2, B_1, B_2 \) for the concentration of nitrogen impurities \( x = 0.08\% \) are \( A_1 = 0.92, A_2 = 0.08 \) and \( B_1 = -0.55 = -B_2 \). On the other hand, we will also need approximate expressions for large \( B \),

\[
E_{n_1} = E_L - \frac{2x_C^2}{n_1 + \frac{1}{2}} (\hbar \omega_c) + O \left( \frac{1}{(\hbar \omega_c)^2} \right), \tag{12a}
\]

\[
E_{n_2} = \left( n_2 + \frac{1}{2} \right) \hbar \omega_c + \frac{2x_C^2}{n_2 + \frac{1}{2}} (\hbar \omega_c) + O \left( \frac{1}{(\hbar \omega_c)^2} \right). \tag{12b}
\]

It is obvious that the lower branch \( E_{n_2} \) pins to the nitrogen induced level, \( E_L \), for large magnetic fields. Furthermore, there is a gap between the values for \( B \to 0 \) and \( B \to \infty \) which is given by

\[
\delta_0 = \frac{1}{2} \left( \sqrt{E_L^2 + 4x_C^2} - E_L \right). \tag{13}
\]

This is a consequence of the fact that we have a two-level like problem whose energy separation is determined by the matrix element \( C_N \), and the concentration \( x \). For instance, for \( x = 0.08\% \), its value is \( \delta_0 = 23\text{meV} \).

Raman measurements\cite{14} indicate that the localization radius lies in the range \( a_{HW} = 1.25 - 1.7\text{nm} \) \( (a_{HW} = a\sqrt{n}\) \), so that the parameter \( a \) is of order 2nm. For the present model to be valid, one should impose the condition that the cyclotron orbit is at least three times larger than the spatial extent of the impurity wavefunction \( |\psi_L|^2 \). This ensures that the magnetic field does not distort the impurity wavefunction, and it yields the upper bound \( B_{\text{max}} = 35\text{T} \) for the magnetic field. The coefficients \( \alpha \) and \( \beta \) are found from their ratio (determined by the corresponding eigenvalue equation) and the condition that the wavefunction be normalized (the phase factors of \( \alpha \) and \( \beta \) are taken to be zero):

\[
\frac{\alpha_{n_1}}{\beta_{n_1}} = \frac{\sqrt{x_CN}}{E_{n_1} - E_{Cn_1}}, \quad \alpha_{n_1}^2 + \beta_{n_1}^2 = 1, \tag{14}
\]

which results into

\[
\alpha_{n_1} = \frac{\sqrt{x_CN}}{\sqrt{x_C^2N + (E_{n_1} - E_{Cn_1})^2}}, \tag{15}
\]

\[
\beta_{n_1} = \frac{E_{n_1} - E_{Cn_1}}{\sqrt{x_C^2N + (E_{n_1} - E_{Cn_1})^2}}. \tag{16}
\]

These expressions are essentially the same as those of Ref.\cite{15}. The overlap between the localized impurity wavefunction and conduction band states is neglected which is justified for \( B < B_{\text{max}} = 35\text{T} \).

### III. Electron-Phonon Interaction in GaAs:N

In order to obtain more precise values for the energy levels, one must also take into account the interaction of electrons with the thermal vibrations of the crystal. In a polar semiconductor like GaAs, electrons interact with longitudinal optical (LO) phonons more strongly than with other types of phonons. They may be assumed dispersionless having energy \( \omega_{LO} = \hbar \omega_{LO} = 36\text{meV} \) in GaAs\cite{16}. Bearing in mind that coupling with the LO phonons is weak in common semiconductors, Fröhlich\cite{17} proposed the following form of the Hamiltonian

\[
H = H_0 + \hbar \omega_{LO} \sum_q b_q^\dagger b_q + H_{e-ph} \tag{17}
\]

where the expression for the interaction part with the LO phonons reads

\[
H_{e-ph} = \sum_q V_q e^{i\mathbf{q}\cdot\mathbf{r}} (b_{q}^\dagger + b_{-q}), \tag{18}
\]

with

\[
V_q^2 = \frac{4\pi\alpha\hbar(\hbar \omega_{LO})^{3/2}}{(2m^*)^{1/2}\Omega q^2}. \tag{19}
\]

The strength of the electron-phonon interaction depends on the dimensionless coupling constant \( \alpha \) which for GaAs has a small value of\cite{16} 0.068. The interaction becomes important when the cyclotron energy \( \hbar \omega_c \) approaches the energy of the longitudinal optical phonon \( \hbar \omega_{LO} \). This corresponds to the situation when, for instance, the energy of the unperturbed ground state with one real phonon \( |n = 0, 1_{ph} \rangle \), \( E_0^{(0)} = \hbar \omega_{LO} + 1/2 \hbar \omega_c \) crosses\cite{16} the first excited Landau state with no phonons \( |n = 1, 0_{ph} \rangle \), \( E_1^{(0)} = (3/2)\hbar \omega_c \). The electron-phonon interaction removes the degeneracy at \( \omega_c = \omega_{LO} \) and makes the two level anticross. The renormalized values \( E_0 \) and \( E_1 \) of the two levels can be found using perturbative methods. However, the usual Rayleigh-Schrödinger perturbation theory (RSPT) does not give precise values
of the excited states for large values of the magnetic field. To overcome this problem, an improved Wigner-Brillouin perturbation technique [16, 18, 19] (IWBPT) is usually employed to determine the pinning values of the renormalized levels. The energy correction for the \( n \)-th state within this method is given by the expression [16]

\[
\Delta E_n = \sum_{m=0}^{\infty} \sum_{q} \frac{|M_{nm}(q)|^2}{D_{nm}},
\]

and the matrix element \( |M_{nm}|^2 \) has the following form for the first two energy levels

\[
|M_{0m}|^2 = |\alpha_0 \alpha_m|^2 \frac{e^{-s}}{m!} s^m \quad (n = 0),
\]

\[
|M_{1m}|^2 = |\alpha_1 \alpha_m|^2 \frac{e^{-s}}{m!} s^{m-1}(m-s)^2 \quad (n = 1),
\]

where \( s = (q_B l_c)^2/2 \). According to this perturbation scheme, the denominator \( D_{nm} \) depends on the energy correction itself [18], \( \Delta E_n \),

\[
D_{nm} = E_n - E_m (-q_B) - \hbar \omega_{LO} + \Delta E_n - \Delta E_0,
\]

so that the solution must be sought self-consistently. The correction to the ground state energy \( \Delta E_0 \) is equal to its counterpart within Rayleigh-Schrödinger perturbation theory, i.e. \( \Delta E_0 = \Delta E_{0}^{RS} \). Fig. 2 shows the values of the first transition energy \( E_1 - E_0 \) as a function of cyclotron energy \( \hbar \omega_c \) (i.e. magnetic field), by the solid curves. For comparison, the values of the same quantity are given for pure GaAs by the dashed curve.

The anticrossing behavior of the levels \( E_0 \) and \( E_1 \) is obvious. The lower curves, which correspond to the case below the phonon continuum pin to \( \hbar \omega_{LO} \) for large magnetic fields. The difference between GaAs:N and GaAs is the largest when the cyclotron energy is comparable to the LO phonon energy, \( \hbar \omega_{LO} \). This difference should increase as the cyclotron orbit becomes comparable to the localization radius \( l_c \approx a \), but eventually both values should pin to \( \hbar \omega_{LO} \) for very large \( B \). Another important difference between GaAs:N and pure GaAs is that the values of \( E_1 - E_0 \) above the phonon continuum in former (doped) case do not tend to \( \hbar \omega_c \), but to a somewhat smaller slope around \( A_1 \) (see Eq. (10a)). This is the consequence of the nonparabolicity of the conduction band of GaAs:N. The minimum difference between the two levels is \( \Delta E_{10} = 5 \text{meV} \) and it occurs at magnetic field \( B = 23 \text{T} \) just above the longitudinal phonon energy \( \hbar \omega_{LO} \).

IV. CYCLOTRON RESONANCE ABSORPTION

Cyclotron resonance measurements is a standard technique [20] to measure effective masses in bulk semiconductors. In pure semiconductors, when scattering on defects and impurities can be neglected, and for the case of parabolic bands, absorption should ideally consist of a single sharp peak located at the cyclotron energy \( \hbar \omega_c \). However, in case of GaAs:N, it will be shown that the absorption linewidths are naturally broadened due to the change in the conduction band structure caused by the nitrogen impurities. Absorption is a measurable quantity which is determined by the oscillator strength that is defined by [21, 22]

\[
I_{fi} = \frac{2}{m^* E_{fi}} |\langle i | \hat{\rho}_y | f \rangle|^2.
\]

In this work we will consider only transitions between adjacent levels, \( n_1 - 1 \rightarrow n_1 \), that are normally only possible in cyclotron measurements. In the calculation of the matrix elements \( d_{n_1,n_1-1} = \langle n_1 - 1 | \hat{\rho}_y | n_1 \rangle \), only one term survives [13] when the total wavefunction, Eq. (9) is inserted in Eq. (23), and

\[
d_{n_1,n_1-1} = \alpha_{n_1} \alpha_{n_1-1} \frac{\hbar}{l_c} \sqrt{\frac{n_1}{2}},
\]

while for the states higher than the impurity state, \( E_{fi} \), the expression is the same with \( n_1 \) replaced by \( n_2 \). The coefficients \( \alpha_{n_1} \) and \( \alpha_{n_1-1} \) are given by the expression Eq. (15). In the next two figures, Fig. 3 and Fig. 4 we present the oscillator strengths for the first five transitions between adjacent levels \( n - 1 \rightarrow n \), within the lower and upper subbands, as a function of magnetic field \( B \). The values of the oscillator strengths for the lower subbands decrease with increasing magnetic field \( B \) (Fig. 3), due to the presence of coefficients \( \alpha_{n_1} \) and \( \alpha_{n_1-1} \).
in Eq. (24). This can be explained by the fact that conduction like states \( E_{n_1} \) acquire somewhat of a localized nature as they approach the pinning value of \( E_L \). On the other hand, the strengths for upper subbands start from very small values indicating their highly localized nature at small magnetic fields. At the end, it would be useful to calculate the absorption coefficient \( \alpha_{abs} \) as a function of the energy of the incident light, \( \hbar \omega \). Within the dipole approximation, the general formula reads\(^{24}\)

\[
\alpha_{abs}(\hbar \omega) = \frac{\mu_0 c \pi e^2 \hbar}{2m^* n_r} \sum_{i,f} I_{fi} \rho_j(\hbar \omega)(f(E_i) - f(E_f)) \quad (25)
\]

where \( n_r \) is the refractive index, \( \rho_j \) is the joint density of states, and \( \hbar \omega \) is the energy of the incident light. The last factor in Eq. (25) is the difference in the Fermi-Dirac distribution of the initial and the final state \( (E_i, E_f) \). Note that in Eq. (24) one should use the modified (joint) density of states, \( \rho_j \), to take proper account of the presence of N impurities. The sum in the same equation will in practice terminate due to the finite Fermi level and temperature, while in the case of interest \( f = i + 1 \) due to selection rules. In the next figure, Fig. 5 the theoretical estimate for the absorption coefficient for intraband transitions within the lower subband are shown as a function of the incident energy \( E = \hbar \omega \) at room temperature \( T = 300K \) for two different values of magnetic field: \( B = 10T \) (solid curve) and \( B = 20T \) (dashed curve). In both cases the electron concentration was kept constant at \( n_c = 2 \times 10^{17} \text{cm}^{-3} \), so that the position of the Fermi level changes with magnetic field. To achieve better clarity, a break point is introduced on the \( x \)-axis between 16 and 22meV. The broadening and asymmetry of the absorption lines is a consequence of the band nonparabolicity of \( E_{n_1}(k_z) \) introduced by the nitrogen impurities. For the same reason, the local maxima are not located at \( \hbar \omega_c \) but do depend on the quantum number \( n_1 \), since the equation \( E_{n_1}(k_z) - E_{n_1-1}(k_z) = \hbar \omega \) is not trivial.

V. SUMMARY AND CONCLUSIONS

In this work we presented a model to determine the energy levels in dilute nitride GaAs:N under applied magnetic field, taking into account electron-phonon interaction. The model is based on the Band Anticrossing Model for GaAs:N but here modified to include the interaction with the longitudinal optical phonons treated within second order perturbation theory. It is assumed that the impurity wavefunction has a Gaussian shape, spanning just a few lattice constants. The influence of nitrogen impurities are characterized by three parameters: localized level \( E_L \), the matrix element \( C_N \) and concentration of the impurities \( x \). The polaron correction to the energy
levels results in a smaller transition energy than from pure GaAs. This difference is the largest around the LO phonon energy $\hbar \omega_{LO}$, but eventually becomes zero for large magnetic fields. Furthermore it is shown that the oscillator strength for lower subbands (with respect to $E_L$) decrease with increasing of magnetic field as they approach the pinning value of $E_L$. The situation for the upper branch is the opposite, since they acquire somewhat of an extended like nature. This should be revealed in the absorption measurement, in the sense that for only higher magnetic fields the intraband transition within the upper branch may contribute significantly to the absorption spectrum. At the end, the theoretical estimates of the absorption coefficient are done for the lower subband at room temperature $T = 300$K, for two different values of magnetic field $B$. The absorption lines have asymmetric shape due to the non-parabolicity of the conduction band caused by the short range impurity potential.

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