Nonlinear Landau absorption in III-V semiconductors near the fundamental absorption edge

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Analytical investigations have been made of nonlinear Landau absorption in important III-V semiconductors such as GaAs, GaSb, InSb, and InAs which have been irradiated by suitable lasers of photon energies ($\hbar \omega$) nearly equal to the band-gap energies ($\hbar \omega_{\rm r}$) in the presence of a large magnetostatic field. A coherent radiation-exciton interaction model has been used for the direct allowed transitions between the $n = 0$ Landau subbands. The result obtainable for linear magnetoabsorption using the present model agrees very well with that obtained in the 1960's using a completely different approach. The effective nonlinear Landau absorption $\alpha^{nonlin}(B_0)$ has been studied by examination of the imaginary part of the third-order optical susceptibility. The contributions from the higher-order susceptibilities have been neglected. The Wannier-Mott exciton wave function $\psi(0)$ is found to play a very important role in the nonlinear Landau absorption processes as it occurs in the fourth power in the expression for $\alpha^{\text{nonlin}}(B_0)$. For near-band-gap resonant excitation in the true continuum with $\hbar(\omega - \omega_s)$ less than the crystal exciton rydberg, $|\psi(0)| \gg 1$ and its contribution must be recognized under this regime. The renormalization of the crystal band gap due to the exciton rydberg and the light shift has also been discussed.

I. INTRODUCTION

The key role played by the giant nonlinearities in crystal refraction and absorption in achieving optical bistability in semiconductors is well established.¹ The origin of these nonlinear properties lies in the large nonlinear optical susceptibility of the crystals irradiated by a laser with photon energy nearly equal to the band-gap energy. This experimental result has been interpreted differently by different workers. Gibbs et $al.^2$ ascribed it to the saturatio of the excitonic absorption in GaAs while Miller et al ² argued that the excitonic effect is negligible in InSb and that interband resonant transition mechanisms are responsible for the observed giant nonlinearity. The experimental observations of Miller et $al.$ ³ were also explained by Moss⁴ on the basis of the dynamic Burstein-Moss effect. Haug⁵ has discussed how, by a nonperturbative manybody technique which incorporates the long-range Coulomb interaction in the quantum system of the electrons and holes, the nonlinear optical properties of semiconductors could be explained. Kanskaya et al.^{6,7} have claimed the successful observation of the ls Wannier-Mott type of discrete exciton structure of the fundamental absorption edge of InSb and concluded that all of the important III-V semiconductors including GaAs and InSb exhibit the same excitonic structures of the absorption edge. Keeping in view the common nature of the nonlinear optical phenomena and the wide-ranging theoretical interpretations, Sen⁸ proposed an independent model which could explain satisfactorily the experimentally observed results in both GaAs and InSb. Consequently, using the coherent-radiation —exciton interaction model, Sen found reasonable agreement between the theoretically determined value of the third-order susceptibility $\chi^{(3)}$ not only for GaAs and InSb but also for GaSb and InAs.

The same theory was extended later to the case of the above crystals subjected to a very large magnetostatic field to study nonlinear refraction.⁹ Quite interesting, the results⁹ could be expressed in a form identical to that of Wherrett and Higgins¹⁰ (although the approaches are completely different} by neglecting the role of excitons in Ref. 9 and choosing $T_1 = T_2$ in Ref. 10. In this connection, it may be noted that the dephasing time constant T_2 and the population relaxation time constant T_1 are still
regarded as essentially unknown parameters.¹¹ To take regarded as essentially unknown parameters.¹¹ To take into account the numerical contribution of the factor $T_1/T_2 \sim 10^3$, it has been shown⁹ that for very-nearresonant interband transitions in the true continuum near the fundamental absorption edge of the crystals, the contribution of the Wannier-Mott-type exciton wave function could be around 2×10^2 to 5×10^2 for a laser photon energy larger than the crystal band-gap energy by roughly half the exciton Rydberg. From the above discussion, it appears that the coherent radiation-exciton interaction model should also be employed to study nonlinear absorption phenomena in the above class of semiconducting crystals in the presence of a large magnetostatic field. This is possible only if the roles of various nonoptical processes are taken into account such that the optical susceptibihty becomes complex with the imaginary part being responsible for the absorption phenomena. This can be done by introducing phenomenologically a damping parameter into the equation of motion of the probability amplitude of the excited electron-hole pair state. Such an approach is well established in quantum mechanics to take account of damping.

In Sec. II, basic formulations for the perturbational calculation of $\chi^{(3)}$ are presented using the coherent model for direct-gap semiconductors in the presence of a magnetostatic field. Section III deals with the general theoretical formulations which lead to an expression for the intensity-dependent optical susceptibility of the crystals. Section IV is devoted to the comparison of the results of the present analysis for linear magnetoabsorption with those of earlier workers.¹² The phenomenon of nonlinear Landau absorption has been investigated analytically in Sec. V. SI units have been employed throughout.

II. BASIC FORMULATIONS

We consider that all of the chosen III-V semiconductors possess isotropic, nondegenerate, and parabolic bands and permit only direct allowed transitions between the highest valence band and the lowest conduction band. The crystal is subjected to a magnetostatic field B_0 applied along the z axis and consequently, the crystal energy spectra are given by 13

$$
\epsilon_c = \epsilon_{c0} + (\hbar^2 k_z^2 / 2m_c) + (n' + \frac{1}{2})\hbar\omega_{cc} + g_c \beta B_0 M_J
$$

(1)

$$
\epsilon_v = \epsilon_{v0} - (\hbar^2 k_z^2 / 2m_v) - (n + \frac{1}{2})\hbar\omega_{cv} + g_v \beta B_0 M_J
$$

where the suffixes c and v stand for the conduction and valence bands, respectively. ϵ_{co} (ϵ_{vo}) is the energy at the center of the lowest conduction (highest valence) band. center of the lowest conduction (inglest valence) band $m_{c,v}$, $g_{c,v}$, and $\omega_{cc, cv}$ (= |e $B_0/m_{c,v}$ |) are the effective masses, g factors, and the cyclotron frequencies, respectively. β (=e $\hbar/2m_0$, m_0 being the electron mass) is the Bohr magneton and M_J ($=\pm \frac{1}{2}$) is the spin quantum number; n and n' are the Landau quantum numbers. For direct allowed transitions when $B_0\neq 0$, the selection rules yield $n = n'$ and $k_z = k'_z$. If B_0 is very large, one can assume that only the $n = n' = 0$ Landau subbands participate in the band-to-band transitions. Hence, we define the interband transition frequency from Eq. (1) as

$$
\omega_{k_z} = \omega_g + \Omega_c / 2 + (g_c - g_v) \beta B_0 M_J / \hbar + \hbar k_z^2 / 2m_r , \qquad (2)
$$

where we have taken $\epsilon_c-\epsilon_v=\hbar\omega_{k_z}$, $\Omega_c=\omega_{cc}+\omega_{cv}$, and $m_r^{-1} = m_c^{-1} + m_v^{-1}$

We now consider the equations of motion of the probability amplitudes of the crystal ground state $a(t)$ and the excited electron-hole pair state $b(k_z,t)$. A spatially uniform coherent pump electric field $E(t) = E_0 \cos(\omega t)$ acting through a dipole moment $\mu(k_z)$ excites an electron from the valence band to the conduction band at k_z leaving behind a hole at the same value of k_z . Here, we have assumed $\mu(k_z)$ and $E(t)$ to be parallel to each other. The interaction Hamiltonian (assumed to be to dipole type) for this case is $-\mu(k_z)E(t)$. The band-to-band transitions take place between the states a and b such that the Hamiltonians for the two types of transitions are obtained as

$$
H_{ba} = -\mu_{ba}(k_z)E(t) \text{ and } H_{ab} = -\mu_{ab}(k_z)E(t).
$$
 (3)

The screened Coulomb potential is given by

$$
U(\mathbf{r},t) = \int \frac{d\omega}{2\pi} \sum_{\mathbf{k}} \frac{U_{\mathbf{k}} \exp[i(\mathbf{k}\cdot\mathbf{r}-\omega t)]}{\epsilon_{\parallel}(\omega,\mathbf{k})}, \qquad (4)
$$

where $U_k = -4\pi e^2/k^2$ and r is the electron-hole separa-

tion. $\epsilon_{||}(\omega, \mathbf{k})$ is the excitation-dependent longitudinal dielectric function.

During band-to-band transitions in semiconductors, many electron-hole pairs (excitons) are optically created which screen the Coulomb potential of a given exciton. Under high-power excitation, the interparticle spacing can be comparable to the exciton Bohr radius and the screening becomes so strong that the bound state, being no longer stable, undergoes a Mott transition. This situation can be studied by incorporating many-body effects arising from the long-range Coulomb interaction in the quantum system of electrons and holes.⁵ In this article, we are interested in the exciton effects in III-V semiconductors; we consider a comparatively low density of electron-hole pairs at moderately low excitation intensities well below the Mott transition. Accordingly, we have omitted without sacrificing much accuracy the excitation dependence of screening and chosen $\epsilon_{\parallel}(\omega, k) = \epsilon_1$, the lattice dielectric constant.

Using the rotating-wave approximation, one can write down the equations of motion of $a(t)$ and $b(k_z,t)$ on using Eqs. (3) and (4) as

$$
\dot{a}(t) = i \sum_{k_z} \frac{\mu_{ba}(k_z) E_0}{2\hbar} \exp[-i(\omega_{k_z} - \omega)t] b(k_z, t)
$$
 (5a)

and

$$
\dot{b}(k_z,t) + \frac{i}{\hbar} \sum_{k_z'} \langle k_z | U | k_z' \rangle \exp[i(\omega_{k_z} - \omega_{k_z'})] t_b(k_z',t)
$$

= $i \frac{\mu_{ab}(k_z) E_0}{2\hbar} \exp[i(\omega_{k_z} - \omega)t] a(t) - \gamma b(k_z,t).$

(5b)

In writing Eq. (5a), the assumption has been made that the transitions taking place between the crystal ground state a and the possible excited pair states ranging over all of the possible values of k_z participate in the interaction. The term γ in Eq. (5b) is the phenomenological damping constant and takes into account the effects of various nonoptical processes encountered during the interband optical transition. It may be noted in this connection that the coherent model considers the same damping for both the population (T_1^{-1}) and the dipole moment (T_2^{-1}) . Thus, in this investigation, we assume $T_1^{-1} = T_2^{-1} = \gamma$.

It was pointed out by Letokhov and Chebotayev¹⁴ that coherence effects could be maximum only if the two decay constants T_2 and T_1 are equal to each other. Elci and Rogovin¹⁵ used the validity of this assumption while studying the phenomenon of four-wave mixing and phase conjugation in direct-gap semiconductors. Earlier, Sen⁹ had shown that such an assumption could establish the identity between the results obtained by himself (using the coherent radiation-exciton interaction model) and that of Wherrett and Higgins¹⁰ (using the resonant interband transition model) for nonlinear refraction in an InSb crystal.

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III. INTENSITY-DEPENDENT OPTICAL SUSCEPTIBILITY

In order to investigate the phenomenon of intensitydependent optical susceptibility of semiconducting crystals by using the coherent radiation-exciton interaction model, one has to confine oneself to a particular regime of the exciting-laser-pump intensity. It is well known that the excitons can exist only at low pump intensity. As the intensity increases, the Mott transition becomes more and more dominant and has been carefully avoided in the present work, since we realize fully its importance and complexities which go hand to hand.

We consider the weakly bound electron-hole pairs of the Wannier-Mott type in all of the direct-gap important III-V semiconductors with an exciton wave function represented by⁹

$$
\psi_j(r) = \sum_{k_z} B_j(k_z) \exp(ik_z r) , \qquad (6)
$$

with $B_i(k_z)$ satisfying the equation

$$
(\omega_{k_z} - \omega_j)B_j(k_z) + \hbar^{-1} \sum_{k'_z} \langle k_z | U | k'_z \rangle B_j(k'_z) = 0 , \qquad (7)
$$

with ω_i being the eigenvalue obtainable from the hydrogenic series

$$
\omega_j = \omega_g - \hbar^{-1} \epsilon_{ex}(0)/j^2, \ \ j = 1, 2, 3, \dots \tag{8}
$$

Here, $\epsilon_{\rm ex}(0)=m_e e^4/2\hbar^2 \epsilon^2$ and is the exciton Rydberg for the particular semiconductor.

We now proceed to solve Eqs. (5) by assuming solutions of the form

$$
a(t) = Ae^{-i\Omega t},
$$

\n
$$
b(k_z, t) = \sum_j c_j b_j(k_z) \exp[i(\omega_{k_z} - \omega - \Omega)t].
$$
\n(9)

 c_i can be determined by using Eq. (6) and taking $r=0$:

$$
c_j = -A(\mu_{ba}E_0/2\hbar)\psi_j^*(0)/(\Omega + \omega - \omega_j) . \qquad (10)
$$

Taking the Fourier transform of $\psi_i(r)$ from Eq. (6), we obtain

$$
B_j(k_z) = \frac{1}{(2\pi)^3} \int \psi_j(r) e^{-ik_z r} d^3r
$$
 (11)

and using Eqs. (5) , (9) , and (10) along with the normalization condition

$$
|a(t)|^2 + \sum_{k_z} |b(k_z,t)|^2 = 1,
$$

one finds

$$
|A|^2 = \left[1 + \sum_{j} \frac{\left|\frac{\mu E_0}{2\hbar}\right| |\psi_j(0)|^2}{(\Omega_r + \omega - \omega_j)^2 + (\gamma + \Omega_i)^2}\right]^{-1}
$$
(12)

with Ω (= Ω , +i Ω _i) being obtained as

$$
\Omega = \left| \frac{\mu E_0}{2\hslash} \right|^2 \sum_j \frac{|\psi_j(0)|^2}{\Omega + \omega - \omega_j + i\gamma} \ . \tag{13}
$$

In obtaining Eqs. (12) and (13), we have assumed that $|\mu_{ab}| = |\mu_{ba}| = \mu$. Using the above formulations, one can determine the values of $a(t)$ and $b(k_z,t)$ in terms of the physical parameters of the crystal and the pump laser. Consequently, the ensemble average of the time-dependent dipole moment can be determined by using the relation

$$
\langle \mu(t) \rangle = 2\mu \left[a^*(t) \sum_{k_z} b(k_z, t) \exp(i\omega_{k_z} t) \right]
$$
 (14)

$$
\langle \mu(t) \rangle = -\frac{\mu^2 E(t)}{\hbar} \sum_j \frac{|\psi_j(0)|^2}{(\omega - \omega_{jr}) + i\gamma_r} \left[1 + \sum_j \frac{|\mu E_0/2\hbar|^2 |\psi_j(0)|^2}{(\omega - \omega_{jr})^2 + \gamma_r^2} \right]^{-1}, \tag{15}
$$

where $\omega_{jr} = \omega_j - \Omega_r$ and $\gamma_r = \gamma + \Omega_i$.

Equation (15) is identical with Eq. (14) of Ref. 9 where γ was introduced as a correction to the pump frequency ω replacing ω by $\omega+i\gamma$ with $\gamma\ll\omega$. We would now proceed to calculate the total induced polarization $P(t)$ due to the laser-induced band-to-band transitions in the semiconducting crystals when the crystal is subjected also to a very large magnetostatic field. The role of the magnetic field is actually twofold in the type of transitions we are concerned with. One of these roles has already been discussed while defining the transition frequency ω_{k} by using the conventional energy spectra of the crystals [Eq. (2), Sec. II]. The other role comes into effect during the determination of $P(t)$. It is well known that the magnetostatic field causes the sharpening of the density of states. The number of Landau subbands associated with a given quantum number *n* (for $n = n' = 0$) in the range k_z

I is equal to $eB_0V/(4\pi^2\hbar)\Delta k_z$ (Ref. 13). Here, V is the volume of the crystal and $k_z = \pi/L$, L being the length of the cavity.¹⁶ Taking into account the effect of spin degeneracy, this number becomes $eB_0V/2\pi\hslash L$. Thus, we define $P(t)$ at finite B_0 as⁹

$$
P(t) = \frac{eB_0 V}{2\pi\hbar L} \frac{1}{V} \sum_{k_z} \langle \mu(k_z, t) \rangle . \tag{16}
$$

Here, one should note that the number of dipoles per unit volume has been defined as $N=1/V \sum_{k_x}$ with k_z ranging from 0 to ∞ , remembering that for real direct-gap semiconductors, most of the population is around the center of the first Brillouin zone. One can consequently replace \sum_{k_z} by $L/\pi \int_0^{\infty} dk_z$ such that Eq. (16) can be modified to

$$
P(t) = \frac{eB_0}{2\pi^2\hbar} \int_0^\infty \langle \mu(k_z, t) \rangle dk_z \ . \tag{17}
$$

In writing Eqs. (16) and (17), we have assumed that the transition dipole moment depends upon the wave number k_z . This is quite obvious from the definition

$$
\mu = -er_{ab} \quad \text{with } r_{ab} = ip_{ab}/m_0 \omega_{k_z} \,, \tag{18}
$$

where m_0 is the free-electron mass and p_{ab} is the interband momentum matrix element.

The form of μ represented by Eq. (18) also shows how the interaction Hamiltonian is modified in the presence of the magnetostatic field through the transition frequency ω_{k_2} defined by Eq. (2).

Writing

$$
\mu^2 = -\left|\frac{ep_{ab}}{m_0\omega_{k_z}}\right|^2
$$

and using Eqs. (15) and (17) , we obtain

$$
P(t) = \frac{eB_0}{2\pi^2\hbar^2} \frac{|p_{ab}|^2 E(t)}{m_0^2} \int_0^\infty \frac{dk_z}{\omega_{k_z}^2} \left[\sum_j \frac{|\psi_j(0)|^2}{(\omega - \omega_{jr}) + i\gamma_r} \left[1 + \sum_j \frac{|\mu E_0/2\hbar|^2 |\psi_j(0)|^2}{(\omega - \omega_{jr})^2 + \gamma_r^2} \right]^{-1} \right].
$$
 (19)

Equation (19) gives us the total induced polarization in the presence of the magnetostatic field. The summation over j represents all possible exciton states at $j=1,2,3$, etc. Keeping in view the experimental observations^{6,7} of the 1s Wannier-Mott discrete exciton structure of the fundamental absorption edge of important III-V semiconductors and the smallness of the contributions of exciton states for $j \ge 2$ in the discrete spectrum,¹⁷ one can obtain a much simpler expression for $P(t)$ by omitting the summation over j. Consequently, for $j = 1$, one gets

$$
P(t) = \frac{eB_0}{2\pi^2\hbar^2} \frac{|p_{ab}|^2 E(t)}{m_0^2} \int_0^\infty \frac{dk_z}{\omega_{k_z}^2} \frac{|\psi_1(0)|^2}{(\omega - \omega_{1r}) + i\gamma_r} \left[1 + \frac{|\mu E_0/2\hbar|^2 |\psi_1(0)|^2}{(\omega - \omega_{1r})^2 + \gamma_r^2}\right]^{-1}.
$$
 (20)

It may be noted in this connection that we have restricted ourselves to an intensity of the pump laser which is quite moderate and the existence of weakly bound Wannier-Mott excitons is not screened by the free-carrier generation. Under such a regime of the pump intensity, for a real III-V semiconducting crystal, it may be reasonably assumed that

$$
\frac{|\mu E_0/2\hbar|^2 |\psi_1(0)|^2}{(\omega - \omega_{1r})^2 + \gamma_r^2} \ll 1
$$
 (21)

and

$$
\left[1+\frac{|\mu E_0/2\hbar|^2 |\psi_1(0)|^2}{(\omega-\omega_{1r})^2+\gamma_r^2}\right]^{-1}
$$

= $1-\frac{|\mu E_0/2\hbar|^2 |\psi_1(0)|^2}{(\omega-\omega_{1r})^2+\gamma_1^2}+\cdots$ (22)

The higher-order terms can be neglected in this convergent expansion series and consequently, Eq. (20) reduces to

$$
P(t) = \frac{eB_0}{2\pi^2\hbar^2} \frac{|ep_{ab}|^2 E(t)}{m_0^2} \frac{|\psi_1(0)|^2}{\omega - \omega_{1r} + i\gamma_r} \times \int_0^\infty \frac{dk_z}{\omega_{k_z}^2} \left[1 - \frac{|\mu E_0/2\hbar|^2 |\psi_1(0)|^2}{(\omega - \omega_{1r})^2 + \gamma_r^2}\right].
$$
 (23)

We can also express the induced polarization in a crystal possessing the property of inversion symmetry in the form of an expansion series

$$
P(t) = \epsilon_0 \chi E(t) = \epsilon_0 (\chi^{(1)} + \chi^{(3)} |E|^2 + \dots) E(t) , \qquad (24)
$$

where $\chi^{(1)}$ and $\chi^{(3)}$ are the first- and third-order optical

susceptibilities, respectively. Thus, one can find out the intensity-dependent optical susceptibility of the crystal by using Eqs. (23) and (24). Because of inversion symmetry, $\chi^{(2)}$, $\chi^{(4)}$, etc., are all zero; moreover, we are not interested in these components as they are responsible for various passive optical properties, such as, parametric amplification, second-harmonic generation, etc., $\chi^{(1)}$, $\chi^{(3)}$, $\chi^{(5)}$, etc., account the linear as well as nonlinear refraction and absorption processes in semiconductors. We have neglected the contributions of $\chi^{(5)}$, $\chi^{(7)}$, etc, to the active nonlinear optical processes owing to $\chi^{(3)}$.

Equating the expressions corresponding to the same powers of $E(t)$ in Eqs. (23) and (24), we find

$$
\chi^{(1)} = \frac{eB_0}{2\pi^2\hbar^2\epsilon_0} \frac{|ep_{ab}|^2}{m_0^2} \frac{|\psi_1(0)|^2}{\omega - \omega_{1r} + i\gamma_r} \int_0^\infty \frac{dk_z}{\omega_{k_z}^2} \quad (25)
$$

and

$$
\chi^{(3)} = \frac{eB_0}{2\pi^2 \hbar^2 \epsilon_0} \frac{|e p_{ab}|^2}{m_0^2} \frac{|\psi_1(0)|^4}{\omega - \omega_{1r} + i\gamma_r}
$$

$$
\times \int_0^\infty \frac{dk_z}{\omega_{k_z}^2} \frac{|\mu/\hbar|^2}{(\omega - \omega_{1r})^2 + \gamma_r^2} . \tag{26}
$$

From Eqs. (25) and (26), one notices that both $\chi^{(1)}$ and $\chi^{(3)}$ are complex. The real part of $\chi^{(1)}$ is responsible for the linear refraction of the laser beam while the imaginary part takes account of the linear absorption process within the crystal. The optical nonlinearities are explained by the finite real and imaginary parts of $\chi^{(3)}$ considering that the higher-order nonlinear susceptibilities contribute negligibly. In the following sections, we have restricted ourselves only to the absorption processes. We have discussed the refraction phenomena elsewhere.⁹

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IV. LINEAR MAGNETOABSORPTION

From the imaginary part of $\chi^{(1)}$ in Eq. (25), one can study the phenomenon of linear magnetoabsorption in the study the phenomenon of linear magnetoabsorption in the III-V semiconductors. We write $\chi^{(1)} = \chi_r^{(1)} - i\chi_i^{(1)}$ and obtain from Eq. (25),

$$
\chi_{i}^{(1)} = \frac{eB_{0}}{2\pi^{2}\hbar^{2}\epsilon_{0}} \frac{|ep_{ab}|^{2}|\psi_{1}(0)|^{2}}{m_{0}^{2}} \frac{\gamma_{r}}{(\omega-\omega_{1r})^{2}+\gamma_{r}^{2}} \times \int_{0}^{\infty} \frac{dk_{z}}{\omega_{k_{z}}^{2}}.
$$
 (27)

Using Eq. (2) and performing the integration, one finds

$$
\chi_{i}^{(1)} = \frac{eB_0 |ep_{ab}|^2 |\psi_1(0)|^2}{4\pi \hbar^2 \epsilon_0 m_0^2} \frac{\gamma_r}{(\omega - \omega_{1r})^2 + \gamma_r^2} \left[\frac{2m_r}{\hbar}\right]^{1/2}
$$

$$
\times \frac{1}{[\omega_{\alpha}(B_0)]^{3/2}}, \qquad (28)
$$

where

$$
\omega_g(B_0)\!=\!\omega_g+\Omega_c\,/2\!+\!(g_c\!-\!g_v)\beta B_0M_J/\hslash\ .
$$

The linear magnetoabsorption coefficient is defined as

$$
\alpha^{\text{lin}}(B_0) = \frac{\omega}{\eta_0 c} \chi_i^{(1)} \tag{29}
$$

with η_0 being the crystal background refractive index. On using Eq. (28) , this yields

$$
\alpha^{\text{lin}}(B_0) = \frac{\omega}{\eta_0 c} \frac{e^3 B_0 |p_{ab}|^2 |\psi_1(0)|^2}{4\pi \hbar^2 \epsilon_0 m_0^2} \frac{\gamma_r}{(\omega - \omega_{1r})^2 + \gamma_r^2} \times \left[\frac{2m_r}{\hbar} \right]^{1/2} [\omega_g(B_0)]^{-3/2} . \tag{30}
$$

The linear magnetoabsorption coefficient given by Eq. (30} is, thus, seen to incorporate a few additional effects which are present during the laser-semiconductor interaction in the near-resonant transition regime. These effects are the following: (a} the finite electron-hole Coulomb interaction which contributes a factor of $|\psi_1(0)|^2$ to the linear absorption coefficient as well as modifies the energy band gap from $\hbar \omega_g$ to $\hbar \omega_1 = \hbar \omega_g - \epsilon_{ex}(0)$; (b) the finite dynamic Stark effect resulting in the renormalization of the excitonic-effect-modified band gap $\hbar\omega_1$ to $\hbar\omega_{1r} = \hbar\omega_1 - \hbar\Omega_r$, in addition to a contribution towards the damping parameter γ changing it to $\gamma_r = \gamma + \Omega_i$. It should also be noted that the effect (b) is very small for the range of laser intensity that allows the excitons to exist.

We now try to make a comparison between the results presented above with those already accepted as the standard representation of magnetoabsorption phenomena in solids. Elliott et al , 12 while studying magnetoabsorptic followed a completely different approach with no proper attention being paid to the finite Coulomb interaction between the electron-hole pairs as well as the nature of the exciton wave function and its contribution. If in the

present formulation, we consider $\langle k_z | U | k_z' \rangle = 0$ and $\left|\psi_1(0)\right|^2 = 1$, it can be shown without much difficulty
that ω_1 , gets replaced by ω_{k} , for Ω that ω_{1r} gets replaced $(=\Omega_r+i\Omega_i)\ll\omega,\omega_g,\gamma$. Also, the dependence of the induced dipole moment on k_z is neglected such that in the near-resonant band-to-band transitions, one can replace the expression $|ep_{ab}/m_0\omega_{k_*}|^2$ by $|ep_{ab}/m_0\omega|^2$ for μ^2 . Under these assumptions, the present formulations yield

$$
\chi_1^{(1)} = \frac{e^3 B_0}{2\pi^2 \hbar^2 \epsilon_0} \frac{|p_{ab}|^2}{m_0^2 \omega^2} \int_0^\infty dk_z \frac{\gamma}{(\omega - \omega_{k_z})^2 + \gamma^2} \ . \tag{31}
$$

Assuming $(\omega - \omega_{k_z})^2 \gg \gamma^2$, the properties of the δ func tion can be applied such that on using Eqs. (29) and (31}, the linear magnetoabsorption can be found to be

$$
\alpha^{\text{lin}}(B_0) = \frac{e^3 B_0 |p_{ab}|^2}{2\pi^2 \eta_0 \epsilon_0 c \hbar^2 m_0^2 \omega} \int_0^\infty dk_z \delta(\omega - \omega_{k_z}) \qquad (32)
$$

which on integration becomes

$$
\alpha^{\text{lin}}(B_0) = \frac{\omega}{\eta_0 c} \frac{e B_0}{2\pi \hbar^2 \epsilon_0} \left| \frac{e p_{ab}}{m_0 \omega} \right|^2
$$

$$
\times \left(\frac{m_r}{2\hbar} \right)^{1/2} \left[\omega - \omega_g(B_0) \right]^{-1/2} . \tag{33}
$$

The corresponding equation obtained by Elliott et al. [Eqs. (3.9) of Ref. 12] for $n=0$ can be expressed as

$$
\alpha^{\text{lin}}(B_0) = \frac{\omega}{\eta_0 c} \frac{2eB_0}{\pi \hbar^2 c} \left| \frac{ep_{ab}}{m_0 \omega} \right|^2
$$

$$
\times \left(\frac{m_r}{2\hbar} \right)^{1/2} \left[\omega - \omega_g(B_0) \right]^{-1/2} . \tag{34}
$$

While comparing Eqs. (33) and (34), one should also note that the former is in SI units and the latter is in cgs units.

These equations agree remarkably and prove the validity of the present model in explaining magnetoabsorption in III-V semiconductors due to direct allowed transitions in the near-resonant transition regime.

V. NONLINEAR LANDAU ABSORPTION

In this section, we deal with the phenomenon of nonlinear Landau absorption in the crystals in presence of the near-resonant pump laser and a very large magnetostatic field. We represent the nonlinear phenomena in terms of the third-order optical susceptibility. This may reasonably be termed as the effective nonlinearity due to the fact that the contributions from the higher-order terms are very small. To study the effective nonlinear Landau absorption, we consider the imaginary part of $\chi^{(3)}$ from Eq. (26) given by

$$
\chi_i^{(3)} = \frac{eB_0}{2\pi^2 \hbar^4 \epsilon_0} \left| \frac{e p_{ab}}{m_0 [(\omega - \omega_{1r})^2 + \gamma_r^2]^{1/2}} \right|^4
$$

$$
\times |\psi_1(0)|^4 \gamma_r \int_0^\infty \frac{dk_z}{\omega_{k_z}^4}
$$
 (35)

with

$$
|\mu|^2 = |ep_{ab}/m_0\omega_{k_z}|^2.
$$

We define the effective nonlinear absorption coefficient in presence of the magnetostatic field in SI units as

$$
\alpha^{\text{nonlin}}(B_0) = \frac{\omega}{\eta_0^2 c^2} \chi_i^{(3)} \,. \tag{36}
$$

Using Eqs. (35) and (36), one can finally obtain

$$
\alpha^{\text{nonlin}}(B_0) = \frac{5\Omega_c \omega}{128\pi\epsilon c^2 \hbar^3} \left(\frac{2m_r}{\hbar}\right)^{3/2}
$$

$$
\times \left| \frac{ep_{ab}}{m_0[(\omega - \omega_{1r})^2 + \gamma_r^2]^{1/2}} \right|^4
$$

$$
\times |\psi_1(0)|^4 \gamma_r[\omega_g(B_0)]^{-7/2}, \qquad (37)
$$

 $\omega_{1r}, \omega_{g}(B_0)$ and Ω_c being defined earlier. Since Ω_c $(=\omega_{cc}+\omega_{cv})$ is negative for the crystals, the role of the effective nonlinear Landau absorption is to reduce the total magnetoabsorption.

Equation (37) shows that the finite Coulomb interaction plays a very important role in producing a giant nonlinearity in the magnetoabsorption processes in III-V crystals near the fundamental absorption edge. The role of the exciton wave function is to enhance the nonlinearity to a considerable extent particularly when $\hbar(\omega - \omega_g) \leq \epsilon_{ex}(0)$. This is because the Wannier-Mott exciton wave function occurs in the fourth power in the expression for $\alpha^{\text{nonlin}}(B_0)$ and in the true continuum near the crystal absorption edge with $\hbar(\omega - \omega_g) \leq \epsilon_{ex}(0)$, we find $|\psi(0)|^2 \gg 1$. Although no mention has been made of the specific nature of the exciton wave function its nature can be studied by comparing it with the hydrogenic wave function under the influence of a magnetostatic field. The condition $(\omega - \omega_{1r})^2 \gg \gamma_r^2$ is satisfied if one considers offresonant interband transitions when the nonlinearity reduces to an infinitesimally small constant value with $|\psi(0)|^4 \sim 1$ such that the exciton wave function does not play any significant role. It may also be noted from Eq. (37) that the crystal band-gap frequency ω_{g} appears in the modified form

$$
\omega_{1r} = \omega_{g} - \epsilon_{ex}(0)/\hbar + \Omega_{r}
$$

in the discrete ls state and the phenomenological damping term γ is replaced by $\gamma_r = \gamma + \Omega_i$. For laser intensities that do not cause complete screening of the excitons, and the terms Ω_r , and Ω_i are very small and do not effectively produce any modification. But the excitonic renormalization of the crystal band gap must be recognized when one considers the phenomenon of nonlinear Landau absorption in the crystals due to the near-band-gap resonant laser excitation in the discrete exciton spectrum, $\omega < \omega_g$ with $\hslash \, |\omega - \omega_{\mathbf{g}}| \leq \epsilon_{\text{ex}}(0)$.

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- ¹Optical Bistability, edited by C. M. Bowden, M. Ciftan, and H. R. Robl, (Plenum, New York, 1981).
- ²H. M. Gibbs, S. L. McCall, T. N. C. Venkatesan, A. C. Gossard, A. Passner, and W. Wiegmann, Appl. Phys. Lett. 35, 451 (1979).
- ³D. A. B. Miller, S. D. Smith, and A. Johnston, Appl. Phys. Lett. 35, 658 (1979).
- 4T. S. Moss, Phys. Status Solidi B 101, 555 (1980).
- 5H. Haug, (private communication).
- L. M. Kanskaya, S. I. Kokhanovskii, and R. P. Seisyan, Fiz. Tekh. Poluprovodn. 13, 2424 (1979} [Sov. Phys. Semicond. 13, 1420 (1979}].
- 7L. M. Kanskaya, S. I. Kokhanovskii, R. P. Seisyan, and Al. L. Efros, Fiz. Tekh. Poluprovodn 15, 1854 (1981) [Sov. Phys. Semicond. 15, 1079 (1981)].
- SP. K. Sen, Solid State Commun. 43, 141 (1982).
- 9P. K. Sen, J. Phys. C 16, 2603 (1983).
- ¹⁰B. S. Wherrett and N. A. Higgins, J. Phys. C 15, 1741 (1982).
- ¹¹A. Miller, D. A. B. Miller, and S. D. Smith, Adv. Phys. 30, 697 (1981).
- ¹²R. J. Elliott, T. P. McLean, and G. G. Macfarlane, Proc. Phys. Soc. 72, 553 (1958).
- ¹³S. D. Smith, in Handbuch der Physik, edited by S. Flügge (Springer, Berlin, 1967), Vol. XXXV/2a, pp. ²³⁴—318.
- ¹⁴V. S. Letokhov and V. P. Chebotayev, in Nonlinear Laser Spectroscopy (Springer, Berlin, 1977), pp. ⁷⁴—80.
- ¹⁵A. Elci and D. Rogovin, Phys. Rev. B 24, 5796 (1981).
- ¹⁶R. Loudon, in The Quantum Theory of Light (Clarendon, Oxford, 1973), pp. ¹⁰²—107.
- ¹⁷R. J. Elliott, Phys. Rev. 108, 1384 (1957).