Exciton-polaritons in InP: Magnetoreflectance investigation

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The fine-structure parameters of the free exciton in InP are determined from low-, intermediate-, and high-field magnetoreflectance investigations. First, a reconsideration of the linear Zeeman splitting at the low-field limit allowed us to obtain the g factors of the conduction electron and of the valence-band hole. Second, taking into account both the field and the wave-vector dependences of the exciton levels, a systematic calculation of the reflectance spectra has been performed for a wide range of field. After a close comparison between the experimental data and that calculation, we obtained the following set of valence-band parameters: $\gamma_1=5.22$, $\gamma_2=1.83$, $\gamma_3=2.34$, and $\kappa=0.89$. For the g factor of the electron, we found $g_c=1.23$.

I. INTRODUCTION

In most zinc-blende-type semiconductors, the exciton ground state is eightfold degenerate at the Γ point due to the twofold spin degeneracy of the conduction band (Γ_6^c) and the fourfold degeneracy of the valence band (Γ_8^v). Including the exchange interaction, the Γ_6^c-Γ_8^v exciton splits into the dipole-allowed Γ_5 state and the Γ_3 and Γ_4 states.[1] In the presence of an external magnetic field or other anisotropic perturbation, the behavior of the exciton states are different from each other and the admixture of these states gives rise to new eigenstates of the system and peculiar selection rules.[2–6] The quantitative analysis of the variation of these states, both energy levels, and oscillator strengths, can reveal the fine structure of the exciton.

Magnetoreflectance measurements are suitable experimental methods with which one can simultaneously deal with the variation of the energy levels and the oscillator strengths of the exciton states.[5] In the low-field limit, the variation of the exciton energy levels can be obtained in the framework of the perturbation theory. From the works of Altarelli and Lipari[2] and Cho,[3] one finds useful descriptions of the Zeeman splittings and the diamagnetic shifts of a real exciton in cubic semiconductors, but few experimental investigations supply sufficient information in order to achieve a complete analysis of the fine structure of the exciton.[7–9] In the high-field limit, the adiabatic approximation was proved appropriate. The detailed description has also been worked out by Altarelli and Lipari.[4] Using this theory, Bimberg *et al* have successfully interpreted their high magnetoreflectance spectra and derived the Luttinger parameters in the case of InP and GaAs;[10,11] in these papers the exchange interaction has not been taken into account. Finally, concerning the intermediate-field regime, the solution of the problem has shown a great variety in variational calculations by assuming different trial functions to describe the system under investigation. In the simplest case of an isotropic hydrogenlike system, there has been a lot of interest in finding the field dependence of the energy levels, but the application of the method to a real exciton, an anisotropic hydrogenlike system in semiconductors, has not received enough attention. Ekardt *et al.* have recognized such a situation and proposed a new type of trial function.[5,12] Including the exchange interaction, this calculation has been applied to the analysis of the magnetoreflectance data for the case of GaAs and InP. They have obtained very small exchange energies and resolved a series of Luttinger parameters slightly at variance with the findings of the adiabatic approximation in the high-field cases.[10,11]

Since the reflectance spectroscopy in the vicinity of the frequencies of the exciton resonance must be in the polariton picture where the exciton dispersions are essential, one can improve the accuracy of the determinations by considering the following facts: In the case of a degenerate valence band, a simple model leads to two exciton bands for $\mathbf{K}\|[100]$ ($\mathbf{K}$ being the wave vector of the exciton).[13] The kinetic energy of the center-of-mass motion is then given by $\hbar^2K^2/2M_h$ in the case of the heavy exciton for which the periodic part of the wave function is $|\pm\frac{3}{2},\pm\frac{1}{2}\rangle$ and $\hbar^2K^2/2M_l$ for the light exciton for which the periodic part of the wave function is $|\pm\frac{1}{2},\pm\frac{1}{2}\rangle$.[14] The exciton effective masses M_h and M_l can be derived from the Luttinger parameters. In the presence of the magnetic field, the degeneracies of the exciton states for $\mathbf{K}=\mathbf{0}$ diminish but the resonances always appear as doublets and the exciton dispersion curves are different for different polarizations (σ_+, σ_-, π). Consequently, the exciton-photon interaction gives rise to three-branch polaritons, different for each polarization. To be brief, a complete analysis of the magnetoreflectance data can only be reached after (i) an accurate evaluation of the *field-dependent* energy levels and oscillator strengths, including the exchange interaction, and (ii) a line-shape analysis of the reflectance spectra taking into account the appropriate $\mathbf{K}$-*dependent* exciton bands and the multiple-mode polariton effect. *Both* the field and the $\mathbf{K}$ dependences are functions of the Luttinger parameters; a careful investigation of the problem cannot consider them separately.

In this paper we report a systematic analysis of the magnetoreflectance spectra of InP in the case of low-, intermediate-, and high-field ranges. Since there are about

nine exciton parameters [the effective mass (m_e) and the g factor (g_c) of the conduction electron; the Luttinger parameters (γ_1, γ_2, γ_3, κ, and q); the exchange energy (Δ) and the longitudinal-transverse splitting (E_{LT})] in the description of the fine structure and their field dependence,[15] it is meaningful to remove some of these adjustable parameters by other experimental findings. Recently, we have determined the exchange energies Δ and E_{LT} by performing uniaxial stress experiments on a high-purity epitaxial InP sample[6] and we plan to elucidate the problem of g factors (g_c, κ, and q) from our magnetoreflectance measurements of low-field cases. Moreover, we have reported a variational method to study an isotropic hydrogenlike system under arbitrary magnetic field which well described the field dependence of the nondegenerate exciton states.[16] Therefore, taking the above points into consideration, it is possible to give more accurate values for the remaining exciton parameters (γ_1, γ_2, γ_3) by means of a theory-experience comparison of the magnetoreflectance spectra in intermediate–high-field ranges.

The paper is organized in the following way. Section II presents the theory of the exciton states in the intermediate magnetic field regime, and we derive some general formulation to obtain the eigenenergies and the eigenfunctions by a variational approach somewhat similar to that developed by Ekardt.[12] Next, Sec. III gives a theoretical description of the magnetoreflectance due to two **K**-dependent exciton resonances. After a short note on the experiment in Sec. IV, we present the numerical results in the last part of the paper. First, using the low-field magnetoreflectance data, we determine the g factor of the electron and the hole. Second, the best fit of a systematic search of the magnetoreflectance up to 20 T allows us to determine the Luttinger parameters (γ_1, γ_2, and γ_3) more accurately than in the previous works. Some discussion and comparisons are finally gathered at the end of this part.

II. FORMULATION OF THE EXCITON STATES IN A MAGNETIC FIELD

In the presence of an external magnetic field, the relative motion of the electron-hole pairs can be described within the effective-mass approximation. The corresponding Schrödinger equation can be written as follows:

$$H_{ex}\Psi_{ex}=E\Psi_{ex} , \tag{1}$$

and the Hamiltonian consists of all contributions of the system:

$$H_{ex}=H_e\left[\mathbf{p}+\frac{e}{c}\mathbf{A}\right]-H_h\left[-\mathbf{p}+\frac{e}{c}\mathbf{A}\right]-\frac{e^2}{\epsilon|\mathbf{r}|}+H_{exch} , \tag{2}$$

where $\mathbf{r}$ and $\mathbf{p}$ are, respectively, the relative coordinate and its conjugate momentum. $\mathbf{A}$ is the magnetic vector potential and we choose the gauge $\mathbf{A}=\frac{1}{2}\mathbf{B}\times\mathbf{r}$; $-e^2/\epsilon|\mathbf{r}|$ is the Coulomb interaction with the static dielectric constant ϵ; and the last term denotes the exchange interaction. The explicit Hamiltonian of the electron and of the hole, H_e and H_h, respectively, is given by

$$H_e(k)=\frac{\hbar^2k^2}{2m_e}\pm g_c\mu_B B \tag{3}$$

and[17]

$$-H_h(k)=\frac{\hbar^2}{m_0}\left[(\gamma_1+\tfrac{5}{2}\gamma_2)\frac{k^2}{2}-\gamma_2(k_x^2J_x^2+\text{c.p.})-2\gamma_3(\{k_xk_y\}\{J_xJ_y\}+\text{c.p.})-\frac{e}{c}\kappa\mathbf{J}\cdot\mathbf{B}-\frac{e}{c}q(J_x^3B_x+\text{c.p.})\right] , \tag{4}$$

where m_e and g_c are the effective mass and g factor of the electron; γ_1, γ_2, γ_3, κ, and q are the Luttinger parameters; and m_0 is the free-electron mass; J_x, J_y, and J_z are the (4×4) angular momentum matrix for a spin $\frac{3}{2}$ state; $\{xy\}=(xy+yx)/2$; and c.p. means the cyclic permutation of the preceding term.

Neglecting the exchange interaction, the exciton state for zero field has eightfold degeneracy. So the wave function in Eq. (1) should be an eight-component function. Using the Bloch functions of the spin $\frac{1}{2}$ of the electron and the spin $\frac{3}{2}$ of the hole as the representation basis, e.g., $|i\rangle=|m_h,m_s\rangle\equiv|\frac{3}{2},m_h\rangle|\frac{1}{2},m_s\rangle$ ($m_h=\pm\frac{3}{2},\pm\frac{1}{2}$; $m_s=\pm\frac{1}{2}$), the exciton states in the magnetic field can be written as follows:

$$|\Psi_{ex}\rangle=\sum_i\psi_i(r)|i\rangle \quad (i=1,2,\ldots,8) , \tag{5}$$

where we have separated the center-of-mass motion and replaced it in the next section. Thus Eq. (1) is actually a 8×8 system of equations with the eight envelope functions $\{\psi_i(r)\}$. To resolve this eigenproblem, we consider a general case of arbitrary magnetic field and choose the following representation for the spin-$\frac{3}{2}$ state:

$$J_x=\begin{bmatrix}0&\sqrt{3}/2&0&0\\ \sqrt{3}/2&0&1&0\\ 0&1&0&\sqrt{3}/2\\ 0&0&\sqrt{3}/2&0\end{bmatrix} , \tag{6a}$$

$$J_y=\begin{bmatrix}0&-\sqrt{3}/2i&0&0\\ \sqrt{3}/2i&0&-i&0\\ 0&i&0&-\sqrt{3}/2i\\ 0&0&\sqrt{3}/2i&0\end{bmatrix} , \tag{6b}$$

$$J_z=\begin{bmatrix}\frac{3}{2}&0&0&0\\ 0&\frac{1}{2}&0&0\\ 0&0&-\frac{1}{2}&0\\ 0&0&0&-\frac{3}{2}\end{bmatrix} . \tag{6c}$$

The exciton Hamiltonian can be then expressed as

$$H_{\rm ex}=\begin{bmatrix} H_0+H'+g_c\mu_B BI & 0 \\ 0 & H_0+H'-g_c\mu_B BI \end{bmatrix}+H_{\rm exch} , \tag{7}$$

where H_0 and $H'(=H_1+H_2)$, respectively, are the (4×4) diagonal matrix and nondiagonal matrix and I is the unitary matrix. Taking the effective Rydberg, $R_0^*=\mu_0 e^4/2\hbar^2\epsilon^2$ as a unit of energy, the exciton radius, $a_0^*=\epsilon\hbar^2/\mu_0 e^2$ $(1/\mu_0=1/m_e+\gamma_1/m_0)$ as a unit of length, and using a reduced magnetic field parameter $\gamma=e\hbar B/2\mu_0 cR_0^*$, the above Hamiltonians show the following forms:

$$H_0=\begin{bmatrix} H_h & 0 & 0 & 0 \\ 0 & H_l & 0 & 0 \\ 0 & 0 & H_l & 0 \\ 0 & 0 & 0 & H_h \end{bmatrix} , \tag{8}$$

$$H_1=-\sqrt{3}\mu_0(\gamma_2+\gamma_3)/2\begin{bmatrix} 0 & 0 & \Pi_-^2-\zeta\Pi_+^2 & 0 \\ 0 & 0 & 0 & \Pi_-^2-\zeta\Pi_+^2 \\ \Pi_+^2-\zeta\Pi_-^2 & 0 & 0 & 0 \\ 0 & \Pi_+^2-\zeta\Pi_-^2 & 0 & 0 \end{bmatrix} , \tag{9}$$

$$H_2=2\sqrt{3}\mu_0\gamma_3\begin{bmatrix} 0 & \Pi_- p_z & 0 & 0 \\ \Pi_+ p_z & 0 & 0 & 0 \\ 0 & 0 & 0 & -\Pi_- p_z \\ 0 & 0 & -\Pi_+ p_z & 0 \end{bmatrix} , \tag{10}$$

where

$$\begin{aligned} H_{h,l}=&(\mu_0/\mu_{h,l}^{\perp})[p^2+(\gamma^2/4)(x^2+y^2)]-2/r \\ &+\mu_0(1/\mu_{h,l}^{\parallel}-1/\mu_{h,l}^{\perp})p_z^2 \\ &+\gamma\mu_0(2/m_e-1/\mu_{h,l}^{\perp})L_z-2\gamma\mu_0(\kappa J_z+qJ_z^3) \end{aligned} \tag{11}$$

and

$$\Pi=i\nabla+\gamma/2(\hat{\mathbf{B}}\times\mathbf{r}) , \tag{12}$$

$$\Pi_{\pm}=\Pi_x\pm i\Pi_y . \tag{13}$$

In Eqs. (9) and (11), we have introduced an anisotropy factor $\zeta=(\gamma_3-\gamma_2)/(\gamma_3+\gamma_2)$ and used four effective-mass parameters:[12]

$$1/\mu_{h,l}^{\perp}=1/m_e+(\gamma_1\pm\gamma_2)/m_0 , \tag{14a}$$

$$1/\mu_{h,l}^{\parallel}=1/m_e+(\gamma_1\mp2\gamma_2)/m_0 . \tag{14b}$$

Concerning the exchange interaction, the resulting configuration has been given in a phenomenological way:[14]

$$H_{\rm exch}=\tfrac{1}{8}\Delta(3-4\mathbf{J}\cdot\mathbf{S})+E_{\rm LT}\delta_{J,1}(\delta_{M,0}-\tfrac{1}{3}) , \tag{15}$$

where the exchange energy (Δ) and the longitudinal-transverse splitting ($E_{\rm LT}$) correspond, respectively, to the energy difference between the $J=1$ triplet state (dipole-allowed) and the $J=2$ quintuplet state (dipole-forbidden) and the energy difference between the longitudinal exciton $|1,0\rangle$ and the two transverse ones $|1,\pm1\rangle$. Here, we express $H_{\rm exch}$ in the $|m_h,m_s\rangle$ basis as follows:

$$\begin{array}{cccccccc} |\tfrac{3}{2},\tfrac{1}{2}\rangle & |\tfrac{1}{2},\tfrac{1}{2}\rangle & |-\tfrac{1}{2},\tfrac{1}{2}\rangle & |-\tfrac{3}{2},\tfrac{1}{2}\rangle & |\tfrac{3}{2},-\tfrac{1}{2}\rangle & |\tfrac{1}{2},-\tfrac{1}{2}\rangle & |-\tfrac{1}{2},-\tfrac{1}{2}\rangle & |-\tfrac{3}{2},-\tfrac{1}{2}\rangle \end{array}$$

$$\begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & \delta/4 & 0 & 0 & -\sqrt{3}\delta/4 & 0 & 0 & 0 \\ 0 & 0 & \delta'/2 & 0 & 0 & -\delta'/2 & 0 & 0 \\ 0 & 0 & 0 & 3\delta/4 & 0 & 0 & -\sqrt{3}\delta/4 & 0 \\ 0 & -\sqrt{3}\delta/4 & 0 & 0 & 3\delta/4 & 0 & 0 & 0 \\ 0 & 0 & -\delta'/2 & 0 & 0 & -\delta'/2 & 0 & 0 \\ 0 & 0 & 0 & -\sqrt{3}\delta/4 & 0 & 0 & \delta/4 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix} , \tag{16}$$

where $\delta=\Delta+\frac{1}{3}(l-1)E_{\rm LT}$ and $\delta'=\Delta+\frac{2}{3}(l+2)E_{\rm LT}$. Following the discussion of Bonneville and Fishman[18] the delocalization parameter l is 1 for a Wannier exciton and 0 for a Frenkel exciton, respectively.

Since the exciton wave function is a linear combination of the product of the Bloch functions and the envelope functions of the electron-hole relative motion [Eq. (5)], the main problem at the present time is to find the field dependence of these envelope functions and the intensities of coupling between these functions. From a general consideration, we hope that all $\psi_i(r)$ have a hydrogen-function-like behavior at the low-field limit and a Landau-level asymptotical change in the case of high fields, respectively. It has been shown that such a behavior of the envelope function can be achieved by simply expressing them as the product of a hydrogenic function and of the Landau exponential factor (the lowest Landau state) as a trial function, submitted to a variational treatment.[16] As a first step we establish our basis set taking the symmetry of the system into account. In fact, after we introduced the magnetic field, the system is z-axis rotation invariant, so the z component of the envelope angular momentum (m) is a good quantum number. The system is also invariant under reflection with respect to the origin of the coordinates; this gives a definite parity ($\tilde{\pi}$) to the eigenstate. In addition, we express these trial functions in the parabolic coordinates:

$$\psi_i=\sum_j C_{ij}\chi_j \ , \tag{17}$$

$$\chi_j(\xi,\eta)=e^{-\alpha_i\xi\eta}\phi_j(\beta_i\xi,\beta_i\eta,\varphi) \ , \tag{18}$$

where $e^{-\alpha_i\xi\eta}$ is the Landau exponential factor and ϕ_j are the hydrogen functions, including one variational scaling parameter (β_i). Thus, the wave function χ_j has an anisotropic real-space extension but recovers the hydrogen function when $\alpha_i\to 0$ and $\beta_i\to 1$ (low-field limit $\gamma\to 0$). Including the symmetry argument, $\phi_j(u=\beta_i\xi,\ v=\beta_i\eta,\varphi)$ can be written as follows:

$$\begin{aligned}\phi_j(u,v,\varphi)=&A_j e^{im\varphi}e^{-(u+v)/2n_j}(uv/n_j^2)^{|m|/2}\\&\times[1+(-1)^m\tilde{\pi}\hat{P}_{uv}]\\&\times\mathcal{L}_{N_1+|m|}^{|m|}(u/n_j)\mathcal{L}_{N_2+|m|}^{|m|}(v/n_j) \ ,\end{aligned} \tag{19}$$

where $\hat{P}_{uv}$ is a permutation operator for (u,v) and $\tilde{\pi}$ refers to the parity of the given consideration ($\tilde{\pi}=\pm 1$); $\mathcal{L}_{N_1+|m|}^{|m|}(u/n_j)$ and $\mathcal{L}_{N_2+|m|}^{|m|}(v/n_j)$ are the associated Laguerre polynomials ($N_1,N_2=0,1,2,\ldots$) and $n_j=N_1+N_2+|m|+1$; A_i is a normalization constant. Please note that here the index j stands for an ensemble of quantum numbers m, $\tilde{\pi}$, N_1, and N_2, hence we represent χ_j by the Dirac symbol $|N_1^{\tilde{\pi}},N_2,m\rangle$.

Now let us consider the optically allowed states for the σ_-, σ_+, and π polarizations; we have to use eight of the lowest even-parity envelope functions for each of these polarizations. As a good approximation, in the case of "not-too-high" magnetic fields, one can only use the main term of the expansion (17) as trial functions, i.e., let $C_{ij}=\delta_{ij}$ and $\psi_i=\chi_i$. This considerably simplifies the calculation. Thus, the lowest *even parity* envelope functions for a given z component of the angular momentum are $|0^+,0,m=\text{even}\rangle$ and $|1^+,0,m=\text{odd}\rangle$. Among all of these states, the lowest one is $|0^+,0,0\rangle$. On the other hand, the z component of the total angular momentum of the exciton is

$$M=m+m_h+m_s \ . \tag{20}$$

The optical transitions are defined as $\Delta M=-1$, 0, and $+1$ corresponding, respectively, to the σ_-, π, and σ_+ polarizations. By incorporating the properties of the φ-dependent part of the functions and of the p^2, $\Pi_\pm$, and p_z operators in the matrix elements of the Hamiltonians, for the three polarizations we use the envelope functions given in Table I.

In fact, the elementary envelope functions have the following simple forms:

$$\begin{aligned}|0^+,0,m=\text{even}\rangle=&2A_i(\beta_i/n_i)^{|m|}e^{im\varphi}(\xi\eta)^{|m|/2}\\&\times\exp[-\alpha_i\xi\eta-\beta_i(\xi+\eta)/2n_i] \ ,\end{aligned} \tag{21}$$

$$\begin{aligned}|1^+,0,m=\text{odd}\rangle=&A_i(\beta_i/n_i)^{|m|+1}e^{im\varphi}(\eta-\xi)(\xi\eta)^{|m|/2}\\&\times\exp[-\alpha_i\xi\eta-\beta_i(\xi+\eta)/2n_i] \ .\end{aligned} \tag{22}$$

Writing the Hamiltonians H_0, H_1, and H_2 in the parabolic coordinates, one has to consider the following operators:

$$\begin{aligned}P^2=&-\frac{4}{\xi+\eta}\left[\frac{\partial}{\partial\xi}\left[\xi\frac{\partial}{\partial\xi}\right]+\frac{\partial}{\partial\eta}\left[\eta\frac{\partial}{\partial\eta}\right]\right.\\&\left.+\frac{1}{4}\left[\frac{1}{\xi}+\frac{1}{\eta}\right]\left[\frac{\partial^2}{\partial\varphi^2}\right]\right] \ ,\end{aligned} \tag{23}$$

$$P_z=-i\left[\frac{(\eta-\xi)}{(\eta+\xi)}\left[\frac{\partial}{\partial\xi}+\frac{\partial}{\partial\eta}\right]-\left[\frac{\partial}{\partial\xi}-\frac{\partial}{\partial\eta}\right]\right] \ , \tag{24}$$

$$\begin{aligned}\Pi_\pm=&-i\sqrt{\xi\eta}e^{\pm i\varphi}\left[\frac{2}{(\eta+\xi)}\left[\frac{\partial}{\partial\xi}+\frac{\partial}{\partial\eta}\right]\right.\\&\left.\pm\left[\frac{\gamma}{2}+i\frac{1}{\xi\eta}\frac{\partial}{\partial\varphi}\right]\right] \ .\end{aligned} \tag{25}$$

TABLE I. Connection between the envelope functions ($|N_1^{\tilde{\pi}},N_2,m\rangle$) and the Bloch functions ($|m_h,m_s\rangle$) for the three polarizations.

m_h,m_s	σ_+	π	σ_-
$\frac{3}{2},\frac{1}{2}$	$\|1^+,0,-1\rangle$	$\|0^+,0,-2\rangle$	$\|1^+,0,-3\rangle$
$\frac{1}{2},\frac{1}{2}$	$\|0^+,0,0\rangle$	$\|1^+,0,-1\rangle$	$\|0^+,0,-2\rangle$
$-\frac{1}{2},\frac{1}{2}$	$\|1^+,0,1\rangle$	$\|0^+,0,0\rangle$	$\|1^+,0,-1\rangle$
$-\frac{3}{2},\frac{1}{2}$	$\|0^+,0,2\rangle$	$\|1^+,0,1\rangle$	$\|0^+,0,0\rangle$
$\frac{3}{2},-\frac{1}{2}$	$\|0^+,0,0\rangle$	$\|1^+,0,-1\rangle$	$\|0^+,0,-2\rangle$
$\frac{1}{2},-\frac{1}{2}$	$\|1^+,0,1\rangle$	$\|0^+,0,0\rangle$	$\|1^+,0,-1\rangle$
$-\frac{1}{2},-\frac{1}{2}$	$\|0^+,0,2\rangle$	$\|1^+,0,1\rangle$	$\|0^+,0,0\rangle$
$-\frac{3}{2},-\frac{1}{2}$	$\|1^+,0,3\rangle$	$\|0^+,0,2\rangle$	$\|1^+,0,1\rangle$

After some straightforward manipulations, all the matrix elements of the Hamiltonian can be expressed with the following integrals:

$$J_1^k(a,b)=\int_0^\infty\int_0^\infty \xi^1\eta^k \exp[-a\xi\eta-b(\xi+\eta)]d\xi\,d\eta \tag{26}$$

and

$$Y_1^k(a,b)=\int_0^\infty\int_0^\infty (\xi\eta)^k(\xi+\eta)^{-1} \times \exp[-a\xi\eta-b(\xi+\eta)]d\xi\,d\eta\ , \tag{27}$$

where $a=\alpha_i+\alpha_j$ and $b=1/2(\beta_i/n_i+\beta_j/n_j)$. Furthermore, all these integrals can be derived by some recurrent relations from two initial integrals $J_0^0(a,b)$ and $Y_1^0(a,b)$. The first one (J_0^0) can be expressed by the standard exponential integral $E_i(x)$.[19] The second one can be computed numerically.[20]

Now we are able to calculate all the elements of the Hamiltonian. We consider the following algorithm for the computation procedure. First we calculate the eigenfunctions and the eigenvalues of every diagonal matrix element using a standard variational approach. Next, taking the determined eigenfunctions we evaluate all the off-diagonal matrix elements. In the presence of the off-diagonal terms the solutions of Eq. (1) are found as some mixture of the solutions of the problem when only considering the diagonal terms. Thus, given a magnetic field we finally find the eigenvalues and the eigenfunctions of the exciton.

III. FORMULATIONS OF THE MAGNETOREFLECTANCE NEAR THE EXCITON RESONANCES

In the above section we have displayed a general consideration to obtain the magnetic field dependence of the exciton states in a cubic semiconductor. Detailed calculations show (see next section) that, increasing the field strength, all the exciton levels shift toward high energies due to the diamagnetic effect, and split due to the Zeeman effect and the exchange interaction. Hence, a reflectance spectroscopy of multioscillators is necessary for a systematic study of the line shapes of the experimental reflectance data.

We consider three peculiar polarizations for the normal-incident light. In the Faraday configuration (σ_+ and σ_-), the spectra were measured in the case of circularly polarized light with $\Delta M=\pm1$, respectively. In the Voigt geometry (π), the spectra were collected in the case of linearly polarized light; the electric vector of the light was perpendicular to the direction of the magnetic field. For each of the three polarizations, one has only two optically active transitions, involving the exciton states of $m=0$ envelope functions displayed in Table I. Of course the other $m\neq0$ states can also lead to nonzero contributions on the optical spectra, but this contribution is very small on the very-high-energy side and is out of the scope of this work.

Now let us look for the dispersion law of the propagation modes in the crystal. In the polariton picture, the exciton-photon interaction gives rise to new normal modes of the system, the so-called excitonic polaritons which result from the admixture of excitonlike and photonlike states.[21,22] The exciton-photon coupling can be basically stated by the following expression:

$$c^2K^2/\omega^2=\epsilon(\omega,\mathbf{K})\ , \tag{28}$$

where ω is the frequency, $\mathbf{K}$ is the wave vector of the center-of-mass motion of the exciton, c is the velocity of light in vacuum, and $\epsilon(\omega,\mathbf{K})$ is the ω- and $\mathbf{K}$-dependent dielectric function, determined by the macroscopic electric properties of the medium. In the vicinity of the frequencies of the exciton resonance:

$$\epsilon=\epsilon_\infty+\sum_{i=1}^{2}\frac{4\pi f_i\omega_i^2}{\omega_i^2+(\hbar\omega_i/\underline{m}_i)K^2-\omega^2-i\omega\Gamma_i}\ , \tag{29}$$

where the background dielectric constant ϵ_∞ contains the nondispersive contribution of the other oscillations; ω_i and f_i ($i=1,2$) are, respectively, the frequencies of resonance and the oscillator strengths of the excitons which depend on the applied magnetic field; $\underline{m}_i$ ($i=1,2$) represent the exciton effective masses which characterize the center-of-mass motions. Equations (28) and (29) hold formally for each of the three polarizations ($\sigma_\pm,\pi$); ω_i and f_i can be found from the work of Sec. II (see also below).

The finite values of the exciton effective mass are responsible for the spatial dispersion of the dielectric function. Following Kane[13] and Fishman,[14] we supposed that the exciton bands in the $|m_h,m_s\rangle$ basis can be expressed by

$$E_h=\hbar\omega_h+\hbar^2K^2/2M_h\ , \tag{30a}$$

$$E_l=\hbar\omega_l+\hbar^2K^2/2M_l\ , \tag{30b}$$

where ω_h and ω_l are the field-dependent frequencies of resonance of the heavy exciton ($|\pm\frac{3}{2},\pm\frac{1}{2}\rangle$) and the light exciton ($|\pm\frac{1}{2},\pm\frac{1}{2}\rangle$), respectively; M_h and M_l are the mass constant,[14] which are function of the Luttinger parameters (γ_1, γ_2, and γ_3), and we will neglect their changes with the strength of the magnetic field. We note that, for example, in σ_+ polarization, the optically active exciton states are derived from the $|\frac{1}{2},\frac{1}{2}\rangle$ and $|\frac{3}{2},-\frac{1}{2}\rangle$ electron-hole Bloch functions; therefore, the two masses used in Eq. (29) should be M_l and M_h, respectively. Similarly, in the case of the two other polarizations we will take M_h for $|-\frac{3}{2},\frac{1}{2}\rangle$ and M_l for $|-\frac{1}{2},-\frac{1}{2}\rangle$ in σ_- polarization, and M_l for both $|\frac{1}{2},-\frac{1}{2}\rangle$ and $|-\frac{1}{2},\frac{1}{2}\rangle$ in π polarization. We emphasize this remark about the spatial dispersion effect in magnetoreflectance, because it can provide a useful criterion for the deduced Luttinger parameters from the field-dependence analysis of the exciton levels. In other words, the Luttinger parameters must describe not only the field dependence of the whole exciton levels, but also the $\mathbf{K}$ dependence of each exciton state.

An equivalent description of the oscillator strength is referred to by the longitudinal-transverse splitting, which gives the strength of the exciton-photon interaction. As the optically active transitions occur principally with the following Bloch states: $|\frac{1}{2},\frac{1}{2}\rangle$, $|\frac{3}{2},-\frac{1}{2}\rangle$ (σ_+); $|-\frac{1}{2},\frac{1}{2}\rangle$,

$|1-2,-\frac{1}{2}\rangle$ (π); and $|-\frac{3}{2},\frac{1}{2}\rangle$, $|-\frac{1}{2},-\frac{1}{2}\rangle$ (σ_-), in the presence of the magnetic field and/or the exchange interaction, the mixture of each pair and their envelope functions determines the optical transition intensity. The different state with different confinement of the electron-hole relative motion gives a different oscillator strength. Considering, for instance, the polarization σ_+, let the lowest optically active states, $|\sigma_{+1}\rangle$ and $|\sigma_{+2}\rangle$, be approximately

$$|\sigma_{+i}\rangle \approx a_{i1}(r)\,|\tfrac{1}{2},\tfrac{1}{2}\rangle + a_{i2}(r)\,|\tfrac{3}{2},-\tfrac{1}{2}\rangle \quad (i=1,2)\ , \tag{31}$$

where $a_{i1}(r)$ and $a_{i2}(r)$ are the corresponding envelope functions. Using the transformation relations between the electron-hole and the exciton representations:

$$|\pm\tfrac{3}{2},\mp\tfrac{1}{2}\rangle = \pm\sqrt{3}/2\,|1,\pm1\rangle + \tfrac{1}{2}\,|2,\pm1\rangle\ , \tag{32a}$$

$$|\pm\tfrac{1}{2},\pm\tfrac{1}{2}\rangle = \mp\tfrac{1}{2}\,|1,\pm1\rangle + \sqrt{3}/2\,|2,\pm1\rangle\ , \tag{32b}$$

$$|\pm\tfrac{1}{2},\mp\tfrac{1}{2}\rangle = \pm1/\sqrt{2}\,|1,0\rangle + 1/\sqrt{2}\,|2,0\rangle\ , \tag{32c}$$

one gets

$$|\sigma_{+i}\rangle = \tfrac{1}{2}[(-a_{i1}+\sqrt{3}a_{i2})\,|1,1\rangle + (\sqrt{3}a_{i1}+a_{i2})\,|2,1\rangle]\ . \tag{33}$$

Since only the $|1,1\rangle$ state is optically active for the given polarization, the oscillator strength can be written as

$$f_{+i} = \tfrac{1}{4}\,|-a_{i1}(0)+\sqrt{3}a_{i2}(0)|^2 f_0\ , \tag{34}$$

where f_0 is the oscillator strength of the unperturbed exciton state $|1,1\rangle$. The ratio of the oscillator strength is given by

$$\frac{f_{+1}}{f_{+2}} = \frac{|-a_{11}(0)+\sqrt{3}a_{12}(0)|^2}{|-a_{21}(0)+\sqrt{3}a_{22}(0)|}\ . \tag{35}$$

If the field strength tends to zero and the admixture of the states $|\frac{1}{2},\frac{1}{2}\rangle$ and $|\frac{3}{2},-\frac{1}{2}\rangle$ can be neglected, the ratio is 1:3.[3] The consideration is the same for the polarizations σ_- and π, and the corresponding constant ratios are 1:3 and 1:1, respectively.

To be brief, a dielectric function with two interacting, ω- and **K**-dependent oscillators will be used in the analysis of the exciton reflectance spectra for various magnetic field strengths. One can calculate from (28) and (29) the effective refractive index and, consequently, the reflectivity coefficient of the crystal. For a given polarization, three transverse solutions can be found which give rise to three polariton modes in the material. The calculation of each reflectivity spectrum will be done in this formalism using the uniform exciton free-layer model of Hopfield and Thomas.[21]

IV. EXPERIMENT

The samples used in this work were very-high-purity vapor-phase-epitaxial InP layers grown on a (100)-oriented substrate produced by Royal Signal and Radar Establishment (RSRE), St. Andrews Road, Great Malvern, and kindly provided to us by D. S. Robertson. The carrier concentration of the epilayers were $\sim 10^{13}$ cm^{-3} with very high mobility ($\sim 10^5$ cm^2/V s at 77 K). After a special etching in dilute solution of bromine in methanol, the sample was mounted strain free in a helium-flow exchange-gas cryostat and cooled down to 4 K.

The reflectance spectra were taken under normal incidence using a tungsten wire lamp. A sensitive GaAs photomultiplier has been used behind a Jobin-Yvon monochromator and a standard lock-in system. The resolution of the monochromator was typically set to 0.3–0.5 Å/mm. The magnetic field was produced by a conventional superconducting magnet. The maximum field strength of this magnet was 6 T. In order to measure the polarization-dependent reflectance, the light beam passed through a circular or a linear polarizer before entering into the monochromator.

V. RESULTS AND DISCUSSION

Nine parameters are necessary for the description of the fine structure of the exciton in InP: m_e, g_c, γ_1, γ_2, γ_3, κ, q, Δ, and E_{LT}, it does not seem appropriable to find them accurately and simultaneously from a simple analysis of the low- or high-field-limit magnetoreflectance spectra. Instead of this, we will use some well-defined parameters and then consider separately two procedures in order to find the remaining ones. In fact, the effective mass of the conduction electron, m_e, is the less-discussable parameter since it has been determined from several experiments.[23] The values of the exchange energy Δ and the longitudinal-tranversal splitting E_{LT} are also well established by Mathieu *et al.* from the uniaxial stress investigation on InP epilayers; the numerical values are $\Delta = 0.04 \pm 0.02$ meV and $E_{LT} = 0.17 \pm 0.04$ meV,[6] a finding comparable with the determination of Ekardt *et al.*[5] In addition, the anisotropic g factor of the hole, q, has a very small value and can be neglected in the case of InP. Using the values given above, we now have to study the remaining parameters.

A. Low-field limit: linear Zeeman splitting

The behaviors of the 1s exciton state in the low-field limit can be described in the degenerate perturbation scheme. Both theoretical and experimental treatments have demonstrated the possibility to elucidate the g factor of the electron and the hole from low-field magnetoreflectance data. Following Altarelli and Lipari[2] and Cho,[3] we introduce two effective g factors giving the splitting between the heavy-hole exciton and the light-hole exciton, respectively,

$$\Delta E_{jh} = g_{jh}\mu_B B\ . \tag{36}$$

Consequently, the g-factor parameters of the electron and the hole are given by

$$g_c = \lambda(g_{3/2}-g_{1/2})/2 - (g_{3/2}+g_{1/2})/4 + \tfrac{64}{5}M\gamma_3(\gamma_3-\gamma_2)(\mu_0/m_0)\ , \tag{37}$$

$$\kappa = (g_{3/2}+g_{1/2})/8 + \tfrac{32}{5}M\gamma_2\gamma_3(\mu_0/m_0)\ , \tag{38}$$

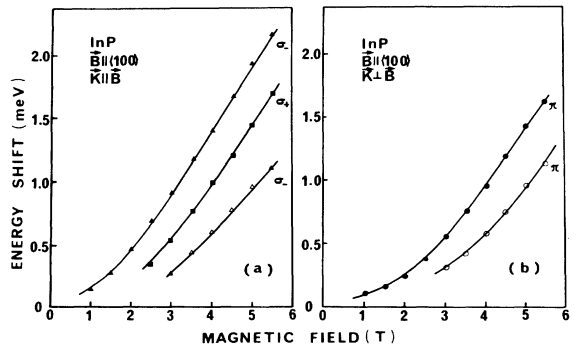

FIG. 1. Observed variation of energy shifts of the reflectance minima of (a) σ spectra and (b) π spectra of the exciton in InP as a function of magnetic field.

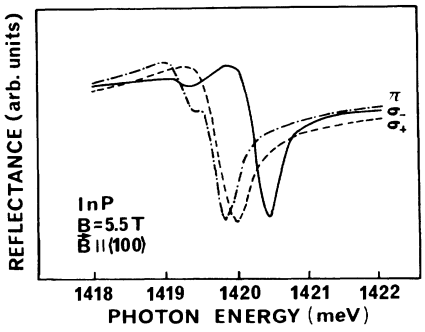

FIG. 2. Typical magnetoreflectance spectra for $B=5.5$ T and for the σ_- (——), σ_+ (— — —), and π (—·—·—·) polarizations.

where $M=0.2812$,[2] and $\lambda=1$, in the case that the exchange energy and the diamagnetic shifts are small enough and the g factors are not appreciably affected.

In Figs. 1(a) and 1(b) we have plotted the energy shifts of the measured reflectance minima as a function of magnetic field for the σ_+, σ_-, and π polarizations. Up to about 3 T ($\gamma\sim0.6$), the strong components reveal a linear Zeeman splitting, and then by increasing the field, a second structure (weak component on the lower-energy side) in the σ_- and π spectra could be observed. Figure 2 shows typical magnetoreflectance spectra for $B=5.5$ T and for the three polarizations. The transverse energies of the exciton states and the energy differences ΔE_{jh} can be determined after an analysis of the line shapes by taking into account the polariton effect and the different dispersions of the exciton states. The effective g factors $g_{3/2}$ and $g_{1/2}$ can be then derived from the fittings in the linear splitting region. We have determined the effective g-factor parameters of the exciton $g_{3/2}=2.4\pm0.2$ and $g_{1/2}=0.35\pm0.2$, which are in agreement with the experimental value deduced by Willmman *et al.*[9] Connecting these data and the Luttinger parameters γ_1, γ_2, and γ_3 of Ref. 5, we get $g_c=1.24\pm0.40$ and $\kappa=0.75\pm0.40$. These values are to be compared with the results obtained by photoluminescence,[28] optical pumping,[30] and two recent magnetoreflectance experiments;[5,10] the agreements are satisfactory (see Table II). On the contrary, concerning the Luttinger parameters γ_1, γ_2, and γ_3, because of the complexity of the splitting and mixing pattern of the energy levels and wave functions, only significant results can be obtained from a detailed analysis of the magnetoreflectance spectra under a sufficiently strong field range.

B. Intermediate field regime: general description

Here we study the variation of the exciton states from magnetoreflectance spectra for a large intermediate-field region. Since only a few of the parameters (γ_1, γ_2, and γ_3) now remain adjustable, their determination will be reliable. Some experimental curves for a range of magnetic field up to 20 T have been recorded by Bimberg *et al.* for all three polarizations.[10] Examples shown in Fig. 3(a) display the behavior of the reflectance spectra for the different field strengths and for the π polarization ($\mathbf{B}\|\langle 100\rangle$ and $\mathbf{E}\|\mathbf{B}$). The polariton structure shifts and increases monotonically with the magnetic field. Again when the field is strong enough, two components in the reflectance appear, which reflect the variation of the energy levels and of the wave functions of the excitons. However, as is well established by the polariton theory, the energy minima E_{mi} of the reflectance curves do not correspond exactly to the exciton energies of resonance E_i. The energy differences $E_{mi}-E_i$ depend on the oscillator

TABLE II. Summary of the band mass parameters and g factors and the exciton effective masses in InP.

g_c	γ_1	γ_2	γ_3	κ	q	M_l	M_h	Reference
	6.73	2.65	2.65			0.20	1.69	24 (calc.)
	6.37	1.39	2.05			0.21	0.45	25 (calc.)
1.20	6.28	2.08	2.76	1.47	0.01	0.22	1.18	26 (calc.)
	5.04	1.56	1.73			0.23	0.83	27 (expt.)
1.15				0.72				28 (expt.)
1.8–2.0				0.7–0.9				9 (expt.)
1.48	5.15	0.94	1.62	0.12		0.25	0.46	29 (expt.)
1.2				0.86($\bar{\kappa}$)				8 (expt.)
1.26								30 (expt.)
1.31	4.95	1.65	2.35	0.97		0.25	1.80	10 (expt.)
1.31	4.95	1.85	2.55	0.97		0.26	10.33	5 (expt.)
1.24	5.22	1.83	2.34	0.89		0.24	1.78	This work

strengths, the damping parameters, and the dead-layer depth, and all of the states vary from each other. *Only a theoretical fit of the experimental spectra allows to get an accurate determination of the exciton states as a function of the magnetic field.*

This is done in Fig. 3(b), where we display the reproductions of the corresponding calculated reflectance spectra. They can be compared with the experimental data of Fig. 3(a). A systematic search based on the theory of Sec. II has been used for the fit. The increase of the amplitude of the structures with the field denotes the increase of the oscillator strength or of the longitudinal-transverse splitting for each exciton state, which have been taken into account automatically by using exciton eigenfunctions. The same comparison between the experimental and calculated reflectance spectra in the vicinity of the exciton resonances for the other polarizations can be performed too. Clearly, as it is shown in Fig. 3(b), the transverse energies of the excitons do not correspond with the energies of the reflectance minima; any attempt to get the fine-structure parameters from a crude estimation of the curve minima may lead to serious errors (the vertical bars E_i indicate the position of the transverse eigenenergies). In Fig. 4(a) and 4(b), the variations of the calculated minima energies have been drawn together with the experimental measurements of Bimberg *et al.* for both Faraday and Voigt configurations. Again, the accordance is satisfactory for this intermediate- and high-field regime. Among the different parameters used in the calculation, most of them were taken from the previous experimental data; the remaining three Luttinger parameters γ_1, γ_2, and γ_3 are introduced as fitting parameters. The best values with the slightly modified κ value are listed in Table II and are compared with the other ones.

Now let us make some additional remarks concerning

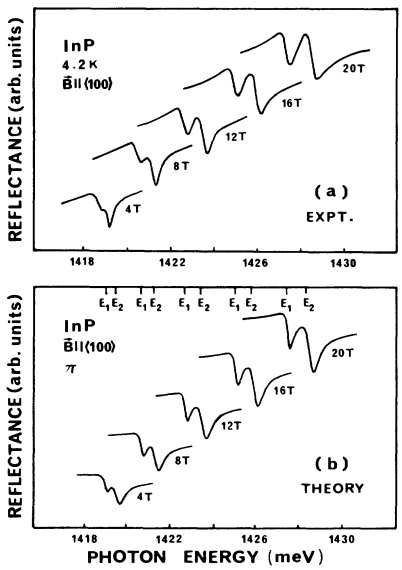

FIG. 3. Magnetoreflectance spectra for various field strengths in π polarization. (a) Experimental spectra obtained at 4.2 K by Bimberg *et al.* (b) Theoretical spectra from a systematic calculation, both the field and the wave-vector dependences of the exciton states have been taken into account. The transverse energies of the exciton are marked.

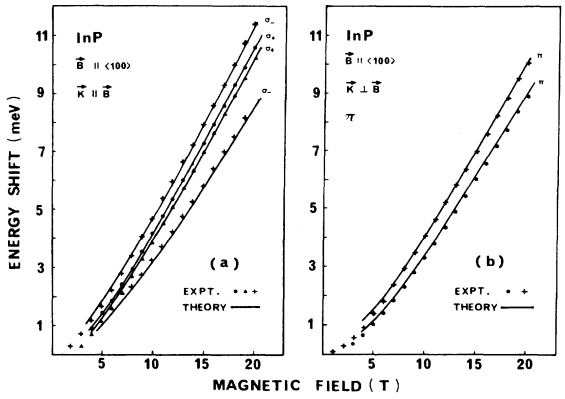

FIG. 4. Variation of energy shifts of the reflectance minima of (a) σ spectra and (b) π spectra of the exciton of InP as a function of magnetic field. Solid lines are present calculations and experimental points are taken from Ref. 10.

this problem.

(i) The exciton in InP can be qualitatively referred to as an anisotropic hydrogenlike system. The degeneracy of the exciton bands gives rise to the linear Zeeman splitting when it is immersed in an external magnetic field. In the absence of the field the relative motion of the electron-hole pair is held only by the Coulomb potential, when the field is introduced the relocalization of the movement is due to both the Coulomb force and Lorentz force. As a consequence, the exciton becomes small in size and the binding energy and the oscillator strength increase. The observable exciton resonances is the effect of the variations of the band structure and the binding energy, but the change of the oscillator strength is more directly related to the field strength.

(ii) The splitting of the energy levels at low fields remains an almost linear function of B, since the magnitude of the exchange interaction is not comparable to the Zeeman splitting in the case of InP. On the other hand, in the intermediate- and high-field range, the mixing of the wave functions are rather sensible to the value of the exchange energy (the longitudinal and transverse splitting too). A comparative study of these parameters favors our earlier finding from uniaxial stress investigations.[6]

(iii) Concerning the Luttinger parameters γ_1, γ_2, and γ_3, the fitting values of this work lead to values in agreement with the results of the most accurate experimental determinations. However, a small discrepancy can lead to a significant difference between the calculated exciton masses M_l and M_h (see Table II) when the parameters of the different findings are used, especially in the case of the heavy-hole exciton one (M_h). These two exciton masses can be directly used for the calculation of the polariton dispersion and of its reflectance spectrum. A more-direct experimental determination of the masses values is required at the present time. Nevertheless, although reflectance spectroscopy is not a most direct method to study the polariton dispersion, the overall agreement for both the Faraday and Voigt configurations leads us to support the present theoretical framework and gives reliable values for the Luttinger parameters.

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