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Free-exciton energy spectrum in GaAs

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The exciton energy spectrum in GaAs has been studied by photoluminescence and reflection. The photoluminescence was excited using a krypton laser (6471 Å). We observed for the first time the spectra for the excited states of the free exciton in GaAs and also the spectra for the ground states in zero field and in the field range from 12 to 40 kG. The value for the exciton binding energy was determined to be $(4.20 \pm 0.3) \times 10^{-3}$ eV from which the band-gap energy of 1.5195 ± 0.0005 eV has been deduced. The values for the Zeeman splitting factors for electron (g factor) and hole (κ parameter) were deduced from these measurements to be $g_c = -0.50 \pm 0.05$ and $\kappa = 1.0 \pm 0.2$, respectively. A phenomenological description for the free-exciton energy spectrum in the intermediate-field regime is used to explain the experimental measurements.

I. INTRODUCTION

A great deal of interest as well as considerable effort has been devoted to understanding the free-exciton energy spectrum of zinc-blende-type semiconductors such as GaAs.[1-9] This work has included both theoretical and experimental investigations both in zero field and with an external magnetic field applied. The ground states of the free exciton have been rather extensively investigated, however the excited states of the exciton have been largely ignored.

The calculations have been carried out in three magnetic field regions: (a) low field, where the magnetic energy is much less than the Coulomb energy; (b) high field, where the magnetic energy is much greater than the Coulomb energy; and (c) intermediate field, where the magnetic energy is of the order of the Coulomb energy. A perturbation scheme[10-12] has been successfully used to explain the observations in the low-field regime. In the high-field regime the adiabatic scheme[10,13-15] has served well. However, in the intermediate-field regime neither of the above schemes can satisfactorily explain the data.

The objectives of this paper are threefold: (a) to present the observation of optical transitions of free excitons in the ground and excited states both in zero field and also in the field regime from 12 to 40 kG, (b) to deduce the various band parameters such as the effective g values for electrons and holes, and (c) to demonstrate that the phenomenological calculation proposed for the exciton energy spectrum in the intermediate-field regime fits the data very well. The value of the exciton binding energy in GaAs of 4.20 ± 0.30 meV has been obtained and this yields the band-gap energy of 1.5195 ± 0.0005 eV. The effective g values for electron and hole are deduced to be $g_c = -0.50 \pm 0.05$ and $\kappa = 1.0 \pm 0.2$.

In Sec. II we discuss the experimental setup and measurements. The theoretical description is discussed in Sec. III. In Sec. IV the various band parameters are deduced and the comparison between the measurements and calculations are discussed in Sec. V.

II. EXPERIMENT

The measurement of magnetic field splitting of free excitons in semiconductors allows one to study the band parameters such as the effective g values for the electron and hole as well as the effective mass and the binding energy of the exciton. In this investigation photoluminescence studies were used to observe the excited states of the exciton while reflection studies were used to observe the ground states. The highest-quality crystals obtainable in GaAs are produced from epilayers. In general, these are thin layers prepared on substrate material and are ideal for photoluminescence studies.

The epilayers used in these experiments have very high mobilities (140 000–250 000 cm^2/V sec at 77 °K). The highest-quality material is needed to observe the excited-state spectra.

The samples were mounted on one end of a sample holder which was in turn placed in the tip of a glass helium Dewar. The mounting was arranged so that the samples were immersed in liquid He. Provision was made for pumping on the liquid He and the temperature was measured by means of vapor-pressure thermometry, using an oil manometer. All of the experiments were conducted in the temperature range 1.2 °K–2.1 °K. The Dewar tip was inserted in the air gap of a conventional dc electromagnet; the maximum field strength of this magnet was 45 000 G. A krypton

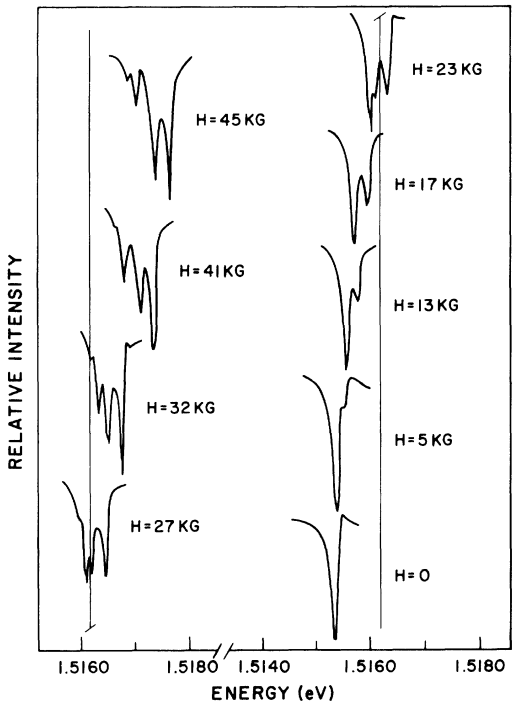

FIG. 1. Reflection intensity as a function of magnetic field.

laser (6471 Å) was used to excite the luminescence. Reflection spectra were taken at near normal incidence using a zirconium lamp. Spectral analysis of the photoluminescence spectra was made with a Bausch and Lomb 2-m grating spectrograph. The spectrograph employed a large high-resolution–diffraction grating producing a reciprocal dispersion of approximately 1.8 Å/mm in first order. With this grating, the spectrographic aperture was about $f/16$. Spectral analysis of the reflection spectra was also made using a 4-m grating spectrograph producing a reciprocal dispersion of 0.9 Å/mm in first order. All of the spectra were photographically recorded on Kodak type 1-N spectroscopic plates. For wavelength calibration, 14 atomic lines from neon (9), vanadium (3), krypton (1), and argon (1) in the range 8161.07 Å–8377.84 Å were used. A wavelength-energy conversion factor of 12395.13 Å eV was used.[2]

The direct exciton at $(K=0)$ is formed by an electron in the conduction band of symmetry Γ_6 $(J=\frac{1}{2})$ and a hole in the valence band of symmetry Γ_8 $(J=\frac{3}{2})$. Excitons formed in this way will have the symmetries $\Gamma_3+\Gamma_4+\Gamma_5=(\Gamma_6\times\Gamma_8)$.

The $j-j$ coupling scheme results in two excitons one with total effective spin $S=1$ and the other with effective spin $S=2$. The $S=1$ and $S=2$ exciton states are split in zero field due to the electron-hole exchange energy. The $S=1$ states are optically allowed in zero field; the $S=2$ states are not. When a magnetic field is present, the $S=2$ states may be optically allowed due to the mixing between the states of $|S=2, m_s=\pm 1, 0\rangle$ and $|S=1, m_s=\pm 1, 0\rangle$.

The exciton ground states as observed in reflection are shown in Fig. 1. These data were taken in the orientation $\vec{K}\perp\vec{H}$, $\vec{E}\perp\vec{H}$. The reflection intensity is plotted as a function of magnetic field strength. Four transitions are observed down to relatively low magnetic field strengths. Linear splittings were observed in the magnetic field region over which the spectra were recorded.

Reflection data were also taken in the orientation $\vec{K}\perp\vec{H}$, $\vec{E}\parallel\vec{H}$. No splitting was observed in these data over the field range investigated. These data are nearly coincident with the data marked C in Fig. 2.

The $n=1$, $n=2$, and $n=3$ states of the free excitons are shown in Fig. 2. The data for the ground-state excitons plotted as solid dots were taken with the 4-m grating spectrograph. The data plotted as open circles were taken with the 2-m grating spectrograph. With this lower dispersion the weak low-energy state was not re-

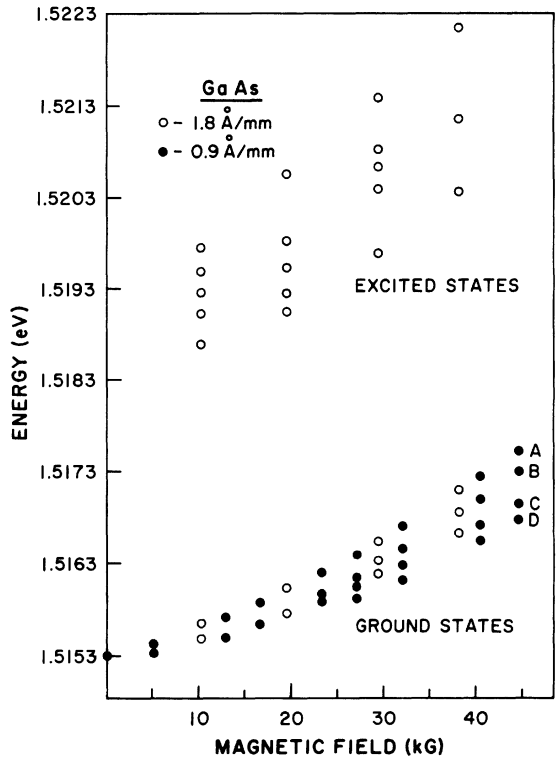

FIG. 2. Experimentally observed ground and excited states of free excitons in GaAs. Energy of the states is plotted relative to the zero-field ground-state energy. The open circles are measured at a dispersion of 1.8 Å/mm; closed circles, at 0.9 Å/mm.

solved. In zero field the ground-state energy is 1.5153 eV and the $n=2$ excited-state energy is 1.5185 eV. These data yield the exciton binding energy of 4.2 ± 0.3 meV and the band-gap energy of 1.5195 ± 0.0005 eV. The ground-state energies were observed in reflection; the excited-state energies were observed in photoemission.

In the presence of a magnetic field, many lines were observed. Some of them, particularly the $n=2$ and $n=3$ states, were observed for the first time.

III. THEORY

In this section we discuss the theoretical description of excitons in semiconductors such as GaAs having zinc-blende-type structures and we present a phenomenological scheme which satisfactorily accounts for the measurements in the intermediate-magnetic-field regime.

The Hamiltonian for the exciton may be written in a form described by Luttinger,[16]

$$H_{ex}=H_e+H_h+H_{eh}, \tag{1}$$

where H_e and H_h are the Hamiltonians for the electron and hole and H_{eh} is the interaction Hamiltonian between the electron and hole,

$$H_{eh}=-e^2/\epsilon|r_e-r_h|+H_{exch}. \tag{2}$$

Here ϵ is the dielectric constant of the medium and H_{exch} is the electron-hole exchange Hamiltonian

$$H_{exch}=A_1\vec{\sigma}\cdot\vec{J}+A_2(\sigma_x J_x^3+\sigma_y J_y^3+\sigma_z J_z^3), \tag{3}$$

where $\vec{\sigma}$ and $\vec{J}$ are the operators for electron spin and effective hole spin, respectively. The parameters A_1 and A_2 are strengths for the exchange energy. This exchange energy lifts the degeneracy of the $|S=1\rangle$ and $|S=2\rangle$ states in zero field.

In general, it is not easy to solve the Hamiltonian of Eq. (1) in the presence of an external magnetic field $\vec{B}$. We discuss it first in the low-field regime, that is, the magnetic energy is much less than the Coulomb energy for the $1s$ ground states. The excited states can be treated in the same way.

The exciton Hamiltonian can be separated into two parts: the spherical symmetric (s-wave-like) and asymmetric (d-wave-like) parts.[17] In the low-field regime one may treat the s-wave-like part as an unperturbed Hamiltonian and the d-wave-like part as an perturbed Hamiltonian.[11] After manipulation,[11,12] the effective Hamiltonian may be written

$$H_{ex}=H_0+H_l+H_q+g_c\mu_B\vec{\sigma}\cdot\vec{B}, \tag{4}$$

where H_0 is the Hamiltonian for exciton in the absence of any external magnetic field, and H_l and H_q are the terms linear and quadratic in magnetic field, respectively. (See the Appendix; hereafter, undefined symbols in the text will be defined in the Appendix.)

We consider the exciton states $|e\rangle$ for our system

$$|e\rangle=\sum_{ij}\alpha_{ij}\left|\tfrac{3}{2},i\right\rangle\left|\tfrac{1}{2},j\right\rangle, \tag{5}$$

where $i=\pm\frac{3}{2},\pm\frac{1}{2}$, $j=\pm\frac{1}{2}$ and α_{ij} are the amplitudes. When a magnetic field, B, in the $\langle001\rangle$ direction is present, we may write immediately the solutions of Eq. (4) as

$$E_{ij}=E_B+G_{ij}\gamma+D_{ij}\gamma^2, \tag{6a}$$

where E_B is the exciton binding energy in the absence of any magnetic field, and γ is the dimensionless magnetic field; G_{ij} and D_{ij} are given in the Appendix.

In the high-field regime, that is, when the magnetic energy is much greater than the Coulomb energy, the solution of Eq. (1) may be obtained by an adiabatic method which can be written as follows[13–15]:

$$E_{ij}=L_{ij}\gamma, \tag{6b}$$

where the L_{ij} are the linear coefficients for the Landau type solutions. They turn out to be of the order of $L_{ij}\approx0.01$. These energies shift much more rapidly with magnetic field than is experimentally observed in the intermediate-field region where our measurements are made.

In the intermediate-field regime, that is, the magnetic energy is of the order of the Coulomb energy, the solutions of Eq. (1) can not be obtained easily. We propose a phenomenological scheme[18] for solutions in this region which accounts well for our data.

The main observations are as follows: (a) in the low-field regime, the eigenvalues should contain the linear Zeeman terms as well as diamagnetic terms and (b) in the high-field regime, the eigenvalues should behave like the Landau level spectra. To satisfy the above observations, a variety of functional forms for eigenvalues can be constructed. In the framework of infinite-order perturbation calculations, one may conclude that the dominant correction term will be an even function of an applied magnetic field, provided the linear Zeeman energy term is absorbed in the unperturbed Hamiltonian. For simplicity, we choose the following form for the eigenvalues for all fields:

$$E_{ij}=E_B+\frac{G_{ij}\gamma+D_{ij}\gamma^2+\beta_{ij}L_{ij}\gamma^3}{1+\beta_{ij}\gamma^2}. \tag{7}$$

In the low-field regime, the above reduces to the perturbation scheme, and in the high-field regime, it reduces to the adiabatic scheme, that is, the Landau level type spectra. The above scheme with $\beta_{ij}=0.5$ accounts very well for the experimen-

tal measurements of the ground-state free exciton spectra in GaAs in the intermediate-field regime, which will be discussed in detail in Sec. V.

IV. CALCULATIONS OF BAND PARAMETERS

We have deduced the values for the effective g factors for electron (g_c), hole (κ), and the exchange energy strength A_1 as follows: Our experiments as well as those of Willmann *et al.*,[7] indicate that the spectra are not very dependent on crystal orientation with respect to magnetic field direction. In view of this we consider our system to be isotropic. It follows that the magnetic-field-dependent Hamiltonian may be written

$$H_B = g(S)\,\vec{S}\cdot\vec{B} + D(S)B^2 ,$$

where $\vec{S} = \vec{J} + \vec{\sigma}$ and the effective g may be given as

$$g(S=1) = -\tfrac{5}{2}\tilde{\kappa} - \tfrac{1}{4}g_c , \quad g(S=2) = -\tfrac{3}{2}\tilde{\kappa} + \tfrac{1}{4}g_c ,$$

and the diamagnetic coefficient $D(S)$ is irrelevant for our purpose since the $D(S)$ is independent of the sign of the magnetic quantum number. When one takes the difference between energies of different magnetic quantum states, the quadratic term in magnetic field is cancelled out.

To complete these calculations unambiguously, level assignments for the data of Fig. 2 must be made. The assignments of the present paper differ from those of Willman *et al.*[7] and are based on the following criteria: (a) applicability of the j-j coupling scheme; (b) observations of circular polarization made by previous investigators[5,7]; and (c) consistent assignment of the $\sigma_\pm$ and π lines for a particular quantum level. The j-j coupling scheme is established from zero-field splitting of an ionized acceptor state observed in our laboratory,[19,20] as well as by line intensity variations described by Willmann *et al.*[7,12] Circular polarization measurements of the $\sigma_\pm$ lines made earlier[5,7] indicate that either line B or line D can be paired with line A for the same value of S. Consistent with the theoretical requirement to have the π line fall between the σ_+ and σ_- lines,[12] the assignment of the same S value to lines A and D follows. Finally, line intensity observations require assignment of $S=1$ to the A line. The assignment of $S=1$ to the A-D pair and $S=2$ to the B-C pair is consistent with expected variations in line intensity which can be expressed in term of the applied magnetic field B as[18]

$$[M(S) \pm \alpha(S)B]^2 ,$$

where $M(S)$ is the matrix element in the absence of a magnetic field, $M(1) \gg M(2) \simeq 0$, and $\pm\alpha(S)$ is the appropriate average value of the dipole moment associated with $m_s = \pm 1$. It seems to us that the argument of saturation of level splitting suggested by Willmann *et al.*,[7] is a consequence of their level assignments. Physically, the splitting of energy levels should be a function of magnetic quantum number, at least over the range of field strengths considered. We note that the argument of saturation can be avoided simply by adopting the level assignments described here.

We determined $g(S)$ from our data by taking differences between energies of the states $m_s = \pm 1$. We obtained the values for g_c and $\tilde{\kappa}$ as follows:

$$g_c = -0.50 \pm 0.05, \quad \tilde{\kappa} = 1.0 \pm 0.2 .$$

The value of g_c is in very good agreement with the calculated value as well as the measured value.[21] The value of $\tilde{\kappa}$ agrees with the measurements of Willmann *et al.*[7] The binding energy of the exciton in GaAs was determined from the energies of the $n=1$ and $n=2$ states using the effective mass approximation. A value of $(4.20 \pm 0.30) \times 10^{-3}$ eV was obtained. From the exciton series limit the band-gap energy is determined to be 1.5195 ± 0.0005 eV. The effective mass of the exciton is $\mu_0 = 0.048 \pm 0.002$.

To estimate the strengths of the exchange energy, A_1 and A_2, we take the difference between the limiting values of the spectra of the $S=1$ and $S=2$ states. We obtained the value of 0.16×10^{-3} eV for the splitting between the $S=1$ and $S=2$ states which yields 0.08×10^{-3} eV for A_1, assuming $A_2 = 0$. A comparison of our data with previous results is shown in Table I.

V. DISCUSSION

Sell *et al.*[2,3] have identified the lines at 1.51515 eV and 1.5183 eV as the exciton lines of $n=1$ and

TABLE I. Comparison between present data and previous results. E_B and E_g are the exciton binding and band-gap energies, respectively. E_{exch} is the exchange energy splitting.

E_B (meV)	E_g (meV)	g_c	$\tilde{\kappa}$	E_{exch} (meV)	Ref.
			1.1 ± 0.1		7 (expt)
		-0.06	1.72		21 (calc)
		-0.48			22 (calc)
4.2 ± 0.2	1519.2 ± 0.2			-0.05 ± 0.05	1–3 (expt)
4.2 ± 0.3	1519.5 ± 0.5	-0.50 ± 0.05	1.0 ± 0.2	-0.16 ± 0.08	present

$n=2$, respectively, and obtained the exciton binding energy of 4.2 ± 0.2 meV. We observed the exciton lines of $n=1$ and $n=2$ at 1.5153 and 1.5185 eV, respectively, and the exciton binding energy of 4.2 ± 0.3 meV. These measurements are in essential agreement. However, for the exchange energy constant, we observed a slightly larger value (-0.16 ± 0.08 meV) than that of Sell *et al.*[2] of -0.05 ± 0.05 meV.

We have compared our measurements with the results from Eqs. (6a) and (6b), using various combinations of the parameters given in Table II, and have found that the agreement between theory and experiment is very poor. Thus we have proposed a phenomenological scheme as described in Sec. III. We compared our measurements with results from Eq. (7), using the values of various parameters given the top row in Table II[22] and those given in the previous section. In Fig. 3, we present the calculated values and experimental data. The agreement is quite good for the ground states.

For the excited states, we found that the direct calculations of the type[23] of Eq. (6a) for the $2s$ and $3s$ states do not give satisfactory results. We reduced the diamagnetic term by 0.08 and were able to obtain good agreement between the calculated and measured results as shown in Fig. 4.

APPENDIX A

The Hamiltonian linear in the magnetic field H_l is given as[11,12]

$$H_l=-2\mu_B[\tilde{\kappa}\,\vec{J}\cdot\vec{B}+\tilde{q}\,(B_xJ_x^3+B_yJ_y^3+B_zJ_z^3)], \quad \text{(A1)}$$

and the term quadratic in the mganetic field H_q is given as

TABLE II. Band parameters in GaAs.

m_e^*	μ_0	μ_1	μ_2	μ_3	ϵ	Ref.
0.066	0.048	0.823	0.148	∞	12.5	a
	0.048	0.444	0.129			b
	0.045	0.400	0.115			c
0.0665	0.044	0.415	0.088			d

[a]R. Kaplan, M. A. Kinch, and W. C. Scott, Solid State Commun. 7, 883 (1969); G. E. Stillman, C. M. Wolfe, and J. O. Dimmock, *ibid.* 7, 921 (1969).
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$$H_q=(ea/2c\mu_0)^2\mu_0\{C_1\vec{B}^2+C_2(\vec{J}\cdot\vec{B})^2 + C_3[B_xB_y(J_x,J_y)+B_yB_z(J_y,J_z)+B_zB_x(J_z,J_x)]\}, \quad \text{(A2)}$$

where

$$(J_i,J_j)=\tfrac{1}{2}(J_iJ_j+J_jJ_i)\ . \quad \text{(A3)}$$

The last term in Eq. (4) is due to the spin momentum interacting with the magnetic field; g_c is the effective g factor of the electron in the conduction state; μ_B is the Bohr magneton; μ_0 is the reduced effective mass of the exciton; a is the Bohr radius of the exciton

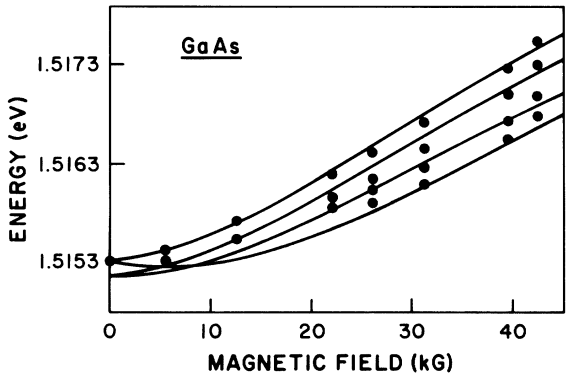

FIG. 3. Energy of the ground-state free excitons as a function of magnetic field. The solid lines are calculated values.

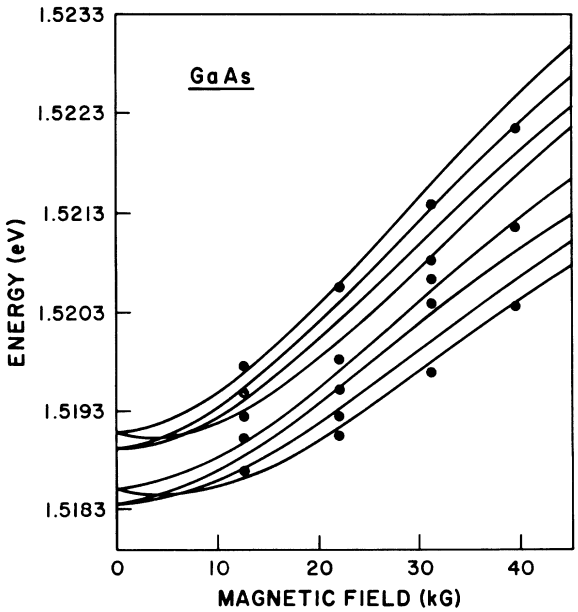

FIG. 4. Energy of the excited states of free excitons in GaAs as a function of magnetic field. The solid lines are calculated values.

$$a = \epsilon/\mu_0 e^2 \ . \tag{A4}$$

Hereafter the units of $\hbar = c = m_0 = 1$, are used. The parameters $\tilde{\kappa}$, $\tilde{q}$, C_1, C_2, and C_3 are related to the Luttinger parameters κ, q, γ_1, γ_2, and γ_3, and also to the effective masses of the conduction electron m_e^* and the free-electron mass m_0, and those of valence-band holes, μ_0, μ_1, μ_2, and μ_3 as follows:

$$\tilde{\kappa} = \kappa - \tfrac{8}{15}(\mu_0/\mu_2)M(1+\tfrac{13}{2}\eta) + \tfrac{14}{3}F_1(m_0/\mu_0)(\mu_0 a v_l)^2 \ , \tag{A5}$$

$$\tilde{q} = q + \tfrac{16}{15}(\mu_0/\mu_2)^2 M\eta - \tfrac{8}{3}F_1(M_0/\mu_0)(\mu_0 a v_l)^2, \tag{A6}$$

$$C_1 = 1 - \tfrac{4}{15}\phi(N+W) - \tfrac{5}{4}C_2 \ , \tag{A7}$$

$$C_2 = (\mu_0/\mu_1)(1+\tfrac{16}{5}W), \tag{A8}$$

$$C_3 = 2C_2\eta + 4F_2(\mu_0 a v_l)^2 \ , \tag{A9}$$

where

$$1/\mu_0 = 1/m_e^* + \gamma_1/m_0 \ , \tag{A10a}$$

$$1/\mu_1 = \gamma_2/m_0 \ , \tag{A10b}$$

$$1/\mu_2 = 2(3)^{1/2}\gamma_3/m_0 \ , \tag{A10c}$$

$$1/\mu_3 = v_l/2m_0\mu_0 \ , \tag{A10d}$$

$$\eta = \tfrac{1}{3}(\gamma_2/\gamma_3 - 1) \ , \tag{A11}$$

$$\phi = 8\gamma_2^2 + 12\gamma_3^2 \ , \tag{A12}$$

$$M = 0.281 \ , \tag{A13a}$$

$$N = 0.469 \ , \tag{A13b}$$

$$W = 0.240 \ , \tag{A13c}$$

$$F_1 = 0.375 \ , \tag{A13d}$$

$$F_2 = 0.844 \ , \tag{A13e}$$

We note that in most cases v_l which is a parameter associated with the energy linear in momentum, is negligibly small.

We consider the exciton states $|e\rangle$ for our system

$$|e\rangle = \sum_{ij} \alpha_{ij} \left|\tfrac{3}{2}, i\right\rangle \left|\tfrac{1}{2}, j\right\rangle \ , \tag{5}$$

where $i = \pm\frac{3}{2}, \pm\frac{1}{2}$, $j = \pm\frac{1}{2}$ and α_{ij} are the amplitudes When a magnetic field, B, in the $\langle 001\rangle$ direction is present, we may write immediately the solutions of Eq. (4) as

$$E_{ij} = E_B + G_{ij}\gamma + D_{ij}\gamma^2 \ , \tag{6a}$$

where E_B is the exciton binding energy in the absence of any magnetic field and

$$G(\pm\tfrac{3}{2}, \pm\tfrac{1}{2}) = \mp(3\tilde{\kappa} + \tfrac{27}{4}\tilde{q})(\mu_0/m_0) \pm g_c \ , \tag{A14}$$

$$G(\pm\tfrac{1}{2}, \pm\tfrac{1}{2}) = \mp(\tilde{\kappa} + \tfrac{1}{4}q)(\mu_0/m_0) \pm g_c \ , \tag{A15}$$

$$D(\pm\tfrac{3}{2}, \pm\tfrac{1}{2}) = \tfrac{1}{2}(C_1 + \tfrac{9}{4}C_2) \ , \tag{A16}$$

$$D(\pm\tfrac{1}{2}, \pm\tfrac{1}{2}) = \tfrac{1}{2}(C_1 + \tfrac{1}{4}C_2) \ . \tag{A17}$$

The unit of energy R^* and the field factor γ are given by

$$R^* = (\mu_0/\epsilon^2)\tilde{R} \ , \tag{A18}$$

$$\gamma = eB/2\mu_0 R^* = (\epsilon/\mu_0)^2(B/B_0), \tag{A19}$$

where

$$\tilde{R} = 13.6058 \text{ eV}, \tag{A20}$$

$$B_0^{-1} = 4.2543\times 10^{-7} \text{ /kG}. \tag{A21}$$

We note that the diamagnetic terms $D(\pm\frac{3}{2}, \pm\frac{1}{2})$ and $D(\pm\frac{1}{2}, \pm\frac{1}{2})$, are different. This causes level crossings at certain values of the applied magnetic field. The only time this is not true is when $C_2 = 0$, in other words, μ_1 is extremely large.

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