Fine structure of excitons and polariton dispersion in quantum wells

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The fine structure of excitons, caused by the electron-hole exchange interaction, is modified in quantum wells due to the reduced symmetry and increased Coulomb interaction. We present a systematic formulation of the exchange interaction for quantum-well excitons and perform numerical calculations of the fine-structure splitting and dispersion for $Al_{0.3}Ga_{0.7}As/GaAs$ quantum wells. The coupling of quantum-well excitons with the electromagnetic field is formulated in second quantization. By solving the coupled equations of motion for excitons and photons we calculate the dispersion relations for localized and resonant polaritons as well as the radiative lifetime for the resonant modes. Our results agree with those obtained by Andreani and Bassani [Phys. Rev. B **41**, 7536 (1990)] and Tassone, Bassani, and Andreani [Nuovo Cimento D **12**, 1673 (1990)], who used Maxwell's equations instead of the quantum field formulation. We draw some conclusions on the fine-structure splitting expected for excitons in quantum wells of II-VI semiconductors.

I. INTRODUCTION

Since the first optical experiments on quantum wells by Dingle, Wiegmann, and Henry[1] it has become clear that excitons play a much more important role in quantum wells (QW) than in bulk material, allowing for the observation of free excitons even at room temperature. This is due to the increased binding energy and oscillator strength of QW excitons, a fact now well understood from the theoretical point of view as a consequence of the increased electron-hole Coulomb interaction resulting from the confinement potential of QW's.[2–5] The fine-structure splitting is caused by the electron-hole exchange interaction and removes the degeneracy of the bound states, which arises from the multiplicity of the conduction- and valence-band edge. Some speculations arose from photoluminescence[6] and magneto-optic experiments[7] with respect to the size of fine structure or exchange splitting of quantum-well excitons. In fact, a simple estimate for two-dimensional (2D) excitons gives an exchange splitting that is orders of magnitude larger than for three-dimensional (3D) excitons.[8,9] However, a theoretical attempt[8] to explain the experimental data of Ref. 6 in a realistic model of QW excitons used a bulk exchange parameter for GaAs which was an order of magnitude larger than the best known experimental values.[9,10] In bulk semiconductors the most accurate experimental information about exchange splitting comes from resonant light scattering[11] and time-of-flight measurements,[12] which at the same time uncover the dispersion of exciton polaritons. In quantum-well systems these experiments are hindered by large exciton linewidths due to well-width fluctuations. However, initial attempts indicate their feasibility in the near future.[13–15] Therefore, a thorough theoretical study of fine structure, dispersion, and polariton effects for quantum-well excitons seems to be in order. The results presented here for $Al_xGa_{1-x}As/GaAs$ quantum wells complement those of Andreani and Bassani[10] and Tassone, Bassani, and Andreani.[16]

In the first part of this paper we will formulate the exchange interaction for excitons in QW's taking into account the light-hole (LH)–heavy-hole (HH) mixing for the subbands as well as the Coulomb coupling between the lowest LH and HH excitons. The exchange splitting of QW excitons, which turns out to depend on the exciton center-of-mass wave vector $\vec{Q}_{\|}$, will be related to the exchange parameters of the bulk excitons. The dominant contribution to the exciton dispersion will be found, however, to derive from the dispersion of the involved electron and hole subbands. In order to evaluate this contribution we exploit the freedom in choosing the relative and center-of-mass coordinates to formulate the hole subband states (and hence the Coulomb interaction) independent of the exciton total in-plane wave vector. Instead it appears in a linear coupling with the relative momentum. This coupling is taken into account in solving the exciton integral equation.

In bulk material the longitudinal-transverse (LT) splitting coincides with the energy shift of the transverse modes if retardation is taken into account and is a direct measure of the interaction strength between excitons and photons. This interaction leads to the concept of excitonic polaritons introduced by Hopfield in 1958, which contributed substantially to our understanding of coupled exciton-photon excitations.[17] In 3D systems it is intimately linked to momentum conservation, i.e., to the fact that each exciton state of a given wave vector couples only to the electromagnetic wave with the same wave

vector and polarization. When transferring this concept from bulk material to a two-dimensional system like a QW, a fundamental difference appears due to the loss of translational symmetry in the growth direction of the sample (taken to be the z direction): the z component of the wave vector is not conserved any more. This induces a coupling between excitons of fixed $\vec{Q}_\parallel$, which is conserved in QW systems, and photons with the same $\vec{Q}_\parallel$ but arbitrary Q_z and leads to two qualitatively different polariton modes: localized (guided) polaritons which have an infinite lifetime and resonant polaritons with a finite lifetime due to radiative decay, as has been pointed out long ago by Agranovich and Dubovskii[18] and in the context of phonon polaritons by Fuchs and Kliewer.[19]

The polariton dispersion has been obtained previously in a semiclassical way by solving Maxwell's equations with a given nonlocal excitonic susceptibility.[10,16,20] Here we give a formulation in the framework of quantum theory. In Sec. IV we derive the Hamiltonian which in second quantization describes the interaction of excitons and photons and solve the corresponding quantum-mechanical equations of motion in Sec. V. The lifetimes of the resonant polaritons, which are related to the imaginary part of the self-energy, are obtained in Sec. VI. Finally in Sec. VII we discuss our results and compare them with existing theoretical and experimental data.

II. THE EXCHANGE HAMILTONIAN

The lowest electronic excitations in a QW system are the excitons formed by an electron in the first conduction subband (C1) and a hole in the first heavy- (HH1) or light- (LH1) hole subband. Taking into account the spin of the particles these exciton states span an eightfold space. The most general form of the exchange Hamiltonian acting in this space can be found by an invariant expansion and was presented in Ref. 9. Due to the reduced symmetry in an $Al_xGa_{1-x}As/GaAs$ QW [the symmetry group for a QW with the growth axis (001) being D_{2d}] as compared to bulk material (symmetry T_d), the exchange Hamiltonian contains a larger number of invariants and, in angular-momentum representation $|M,\sigma\rangle$ falls into four 2×2 blocks corresponding to different values of $|M+\sigma|$ (σ is the electron spin, M the hole spin).[9] The eigenvalues of this exchange Hamiltonian agree with the splitting pattern obtained from the decomposition of $\Gamma_8\otimes\Gamma_6$ (T_d) in the symmetry group D_{2d}, which for HH exciton states is $\Gamma_6\otimes\Gamma_6=\Gamma_1\oplus\Gamma_2\oplus\Gamma_5$ and for LH exciton states $\Gamma_7\otimes\Gamma_6=\Gamma_3\oplus\Gamma_4\oplus\Gamma_5$. The dipole-allowed states are Γ_4 ($\hat{\mathbf{e}}\parallel z$, also called Z exciton) and Γ_5 ($\hat{\mathbf{e}}\parallel x,y$).

Our microscopic approach, which will be presented in the remainder of this section, explains the origin of only a part of the invariants and therefore will not provide the complete splitting pattern. It turns out in this approximation that the dipole-forbidden exciton states, which are of minor interest anyway, will not be affected by the exchange interaction.

The formulation of the Hamiltonian for quantum-well excitons and of the corresponding eigenvalue problem as an integral equation was given by Broido and Sham[2] and Broido and Yang[21] without taking into account the exchange interaction. Here we add the formulation of the exchange contribution. In a first step we evaluate the exchange integral, i.e., the integral over the single-particle wave functions. Its general form is

$$V^X_{\lambda_e\lambda_h\lambda'_e\lambda'_h} = \iint dx_1dx_2\,\psi^\dagger_{\lambda_e}(x_1)\,\psi^\dagger_{\lambda_h}(x_2)\,\frac{e^2}{|\mathbf{r}_1-\mathbf{r}_2|} \times\psi_{\lambda'_h}(x_1)\,\psi_{\lambda'_e}(x_2)\,. \tag{1}$$

The single-particle wave functions for electrons and holes are (the argument $x_{e,h}$ means space and spin coordinates)

$$\psi_{\lambda_e}(x_e) = \sqrt{\frac{\nu}{\mathcal{A}}}\,e^{i\vec{k}_e\cdot\vec{\rho}_e}\sum_{k_z} C(k_z)\,e^{ik_zz_e}\,u_{c\mathbf{k}\sigma}(x_e), \tag{2}$$

$$\psi_{\lambda_h}(x_h) = \sqrt{\frac{\nu}{\mathcal{A}}}\,e^{i\vec{k}_h\cdot\vec{\rho}_h}\sum_{k'_z}\sum_{M} C^j_{\vec{k}_hM}(k'_z)\,e^{ik'_zz_h} \times u_{v\mathbf{k}'M}(x_h)\,, \tag{3}$$

where $C(k_z)$ and $C^j_{\vec{k}_hM}(k'_z)$ are the Fourier transforms of the corresponding subband functions $\zeta(z)$ and $\zeta^j_{\vec{k}_hM}(z)$, $\vec{k}_{e,h},\vec{\rho}_{e,h}$ the in-plane wave vector and particle coordinates, respectively, and $\mathbf{k}=(\vec{k}_e,k_z)$, $\mathbf{k}'=(\vec{k}_h,k'_z)$ the three-dimensional wave vectors. Throughout this paper boldface letters denote 3D vectors whereas 2D vectors in the QW plane are marked with an arrow. λ_e,λ_h are compact notations for the quantum numbers necessary to characterize the single-particle states. $\mathcal{A}$ and $\mathcal{V}$ are the normalization area and volume. $u_{c\mathbf{k}\sigma}(x_e)$ and $u_{v\mathbf{k}'M}(x_h)$ are the spin-dependent periodic parts of the bulk Bloch functions of the electron and hole, which may be constructed from the decomposition of the direct product of spin-1/2 spinors with s- and p-type periodic functions in the symmetry T_d, respectively.[22] They are assumed to be the same for well and barrier material. j labels the four different hole states which due to LH-HH mixing are linear combinations of different M values for the finite in-plane wave vector $\vec{k}_h$. This is accounted for by using the subband $\mathbf{k}\cdot\mathbf{p}$ method which consists of an expansion of the $\vec{k}_\parallel\neq 0$ subband functions in terms of the four series (corresponding to different M values) of bound states at $\vec{k}_\parallel=0$.[23] Within the axial approximation (i.e., the neglect of warping) this expansion can be written as

$$\zeta^j_{\vec{k}_\parallel M}(z) = e^{-i(M-\frac{1}{2})\varphi}\sum_\nu A^j_{\nu M}(k_\parallel)\,\psi_\nu(z)\,, \tag{4}$$

where $\vec{k}_\parallel=(k_\parallel,\varphi)$ in polar coordinates and $\psi_\nu(z)$ is the νth bound state at $\vec{k}_\parallel=0$. The subband functions $\zeta^j_{\vec{k}_\parallel M}(z)$ have definite parity under the operation $z\to -z$, which reduces the number of nonzero expansion coefficients $A^j_{\nu M}(k_\parallel)$.[21] The subband dispersion and the expansion coefficients are obtained by diagonalizing the Luttinger-Hamiltonian including the QW potential.

In performing the integrals in Eq. (1) we first consider the spin part of the Bloch functions and use symmetry arguments to obtain the matrix $(\mathbf{J}\cdot\boldsymbol{\sigma}\text{-}\frac{3}{2})^{\sigma M}_{\sigma' M'}$.[22] In a second step the products of the involved s (p)-type periodic

parts of the electron (hole) Bloch functions are represented as Fourier series

$$u^*_{c\mathbf{k}}(\mathbf{r})\, u_{v\mathbf{k}'}(\mathbf{r}) = \sum_{\mathbf{G}} D_{cv\mathbf{k}\mathbf{k}'}(\mathbf{G})\, e^{i\mathbf{G}\cdot\mathbf{r}}\,, \tag{5}$$

where $\mathbf{G} = (\vec{G}_\parallel, G_z)$ are three-dimensional reciprocal-lattice vectors, in accordance with the assumption that the $u's$ are the same for well and barrier material. By integrating over all space coordinates we obtain

$$V^X_{\lambda_e\lambda_h\lambda'_e\lambda'_h} = -\sum_{M,M'} \tfrac{2}{3}(\mathbf{J}\cdot\boldsymbol{\sigma} - \tfrac{3}{2})^{\sigma M}_{\sigma' M'} \sum_{\mathbf{q}} \frac{4\pi e^2}{\mathcal{V} q^2} \sum_{k_z,\ldots,k'''_z} C^*(k_z)\, C(k'_z)\, C^{j'^*}_{\vec{k}'_\parallel M'}(k'''_z)\, C^{j}_{\vec{k}_\parallel M}(k''_z)$$
$$\times \sum_{\mathbf{G},\mathbf{G}'} D_{cv\mathbf{k}\mathbf{k}''}(\mathbf{G}) D^*_{cv\mathbf{k}'\mathbf{k}'''}(\mathbf{G}')$$
$$\times \mathcal{L}\,\delta_{k_z,k''_z+q_z+G_z}\,\mathcal{L}\,\delta_{k'_z,k'''_z+q_z+G'_z}\,\delta_{\vec{q}_\parallel,\vec{Q}_\parallel-\vec{G}_\parallel}\,\delta_{\vec{q}_\parallel,\vec{Q}_\parallel-\vec{G}'_\parallel}\,, \tag{6}$$

where $\vec{k}_\parallel = \vec{k}_h$, $\vec{k}'_\parallel = k'_h$, $\vec{Q}_\parallel = \vec{k}_e - \vec{k}_h$ is the exciton in-plane center-of-mass wave vector and $\mathcal{L}$ is a periodicity length. In view of the reduced symmetry of the QW system one would expect to find in Eq. (6) instead of the spherical symmetric invariant $\mathbf{J}\cdot\boldsymbol{\sigma}$ the decomposition into $J_x\sigma_x + J_y\sigma_y$ and $J_z\sigma_z$.[9] This is in fact the case, because different weighting factors result in evaluating $D_{cv\mathbf{k}\mathbf{k}'}(\mathbf{G})$ with the in-plane exciton wave vector $\vec{Q}_\parallel$ as a good quantum number, as will be seen in Eq. (10).

In performing the sum over $\mathbf{G}, \mathbf{G}'$ in Eq. (6), we distinguish as in bulk material the contributions for $\mathbf{G} = \mathbf{G}' = \mathbf{0}$ and the rest. In the bulk the former contribution is responsible for the LT splitting of the dipole-allowed Γ_5 exciton states; in the limit $\mathbf{Q} \to \mathbf{0}$ this term depends nonanalytically on the relative orientation between the exciton wave vector $\mathbf{Q}$ and the momentum matrix element $\langle c|\mathbf{p}|v\rangle$. It is called the nonanalytic exchange.[22,24] In contrast the contributions for $\mathbf{G}, \mathbf{G}' \neq \mathbf{0}$ behave analytically. Although it will turn out that for QW's the exciton dispersion is analytic and the LT splitting vanishes linearly for $Q_\parallel \to 0$, we maintain the label "nonanalytic" and "analytic" exchange contribution to give reference to the bulk term from which it derives.

Let us first consider the case $\mathbf{G}, \mathbf{G}' \neq \mathbf{0}$, which corresponds to the analytic exchange contribution. Because $\vec{Q}_\parallel$ and all $\vec{k}'s$ are small compared to the length of reciprocal-lattice vectors, they can be neglected in $D_{cv\mathbf{k}\mathbf{k}'}(\mathbf{G})$ and we have $\mathbf{G} = \mathbf{G}'$. This part of the exchange matrix reads

$$V^{X,A}_{\lambda_e\lambda_h\lambda'_e\lambda'_h} = -\sum_{M,M'} \tfrac{2}{3}(\mathbf{J}\cdot\boldsymbol{\sigma} - \tfrac{3}{2})^{\sigma M}_{\sigma' M'} \sum_{\mathbf{G}\neq\mathbf{0}} \frac{4\pi e^2}{G^2} |D_{cv\mathbf{0}\mathbf{0}}(\mathbf{G})|^2 \frac{\mathcal{L}^2}{\mathcal{V}} \delta_{G_z 0}$$
$$\times \sum_{q_z,k_z} C^*(k_z)\, C^{j}_{\vec{k}_\parallel M}(k_z - q_z) \sum_{k'_z} C(k'_z)\, C^{j'^*}_{\vec{k}'_\parallel M'}(k'_z - q_z)\,. \tag{7}$$

The unknown parameter, which determines the analytic exchange splitting, is given by $A' = \sum_{\mathbf{G}\neq\mathbf{0}} \delta_{G_z 0} \frac{4\pi e^2}{G^2} |D_{cv\mathbf{0}\mathbf{0}}(\mathbf{G})|^2$. At this point we neglect the $\delta_{G_z 0}$ factor and use the bulk parameter $A = \sum_{\mathbf{G}\neq\mathbf{0}} \frac{4\pi e^2}{G^2} |D_{cv\mathbf{0}\mathbf{0}}(\mathbf{G})|^2$, known from the experimental determination of the analytic exchange splitting in GaAs to be $A = 1.98 \times 10^5$ meV.[25] In doing so we obtain an upper limit for this exchange contribution for QW excitons.

The contribution for $\mathbf{G} = \mathbf{G}' = \mathbf{0}$ has to be evaluated by taking into account the differences in the wave vectors $\mathbf{k} - \mathbf{k}'' = \mathbf{k}' - \mathbf{k}''' = (\vec{Q}_\parallel, q_z)$ of the periodic parts of the Bloch functions in a $\mathbf{k}\cdot\mathbf{p}$ expansion, which gives

$$D_{cv\mathbf{k}\mathbf{k}''}(\mathbf{0}) = \frac{1}{\mathcal{V}} \frac{\hbar}{m} \frac{\langle c|(\mathbf{k} - \mathbf{k}'')\cdot\mathbf{p}\,|v\rangle}{E_c - E_v}\,, \tag{8}$$

where c, v refer to the conduction- and valence-band edges of the bulk material. The exchange coupling between different pairs of conduction and valence bands results in a two-band model as a background screening of the exchange term.[24] We obtain for the nonanalytic exchange part

$$V^{X,N}_{\lambda_e\lambda_h\lambda'_e\lambda'_h} = -\sum_{M,M'} \tfrac{2}{3}(\mathbf{J}\cdot\boldsymbol{\sigma} - \tfrac{3}{2})^{\sigma M}_{\sigma' M'} \frac{4\pi\mu^2}{\varepsilon_\infty}\frac{\mathcal{L}^2}{\mathcal{V}} \sum_{q_z} \eta(\vec{Q}_\parallel, q_z) \sum_{k_z} C^*(k_z)\, C^j_{\vec{k}_\parallel M}(k_z - q_z) \sum_{k'_z} C(k'_z)\, C^{j'*}_{\vec{k}'_\parallel M'}(k'_z - q_z)\,, \quad (9)$$

where for GaAs bulk material $\mu = \frac{e\hbar}{mE_g}\langle c|p_x|x\rangle = 26.23\ (\mathrm{eV\,\AA}^3)^{1/2}$, $\varepsilon_\infty = 10.9$ in accordance with a LT splitting of 0.08 meV,[11,25] and for $\vec{Q}_\parallel \parallel \hat{\mathbf{e}}_x$

$$\eta(\vec{Q}_\parallel, q_z) = \begin{cases} \dfrac{q_z^2}{Q_\parallel^2 + q_z^2} & \text{for } |v\rangle = |z\rangle \\ \dfrac{Q_\parallel^2}{Q_\parallel^2 + q_z^2} & \text{for } |v\rangle = |x\rangle \\ 0 & \text{for } |v\rangle = |y\rangle\,. \end{cases} \quad (10)$$

The dependence of $\eta(\vec{Q}_\parallel, q_z)$ on the relative orientation between $(\vec{Q}_\parallel, q_z)$ and the momentum matrix element $\langle c|\mathbf{p}|v\rangle$ gives rise to the longitudinal-transverse splitting. At this point we emphasize that the k dependence of the periodic parts of the electron and hole Bloch functions is crucial for this part of the exchange interaction. Andreani and Bassani[10] in their evaluation of the electron-hole exchange interaction have neglected this k dependence and used a calculation scheme adequate for the description of Frenkel excitons which involves an approximate evaluation of lattice sums.[26] This scheme implies a downward shift of the transverse exciton, which may be interpreted as a local-field correction and is expected for localized Frenkel excitons. This local-field correction was neglected in Ref. 10, in order to achieve a correct description for a delocalized Wannier exciton.

III. DISPERSION OF EXCITONS

The exchange interaction obtained in Sec. II is now considered by first-order perturbation theory. We use the exciton functions calculated within the axial model, which can be written as[3]

$$\phi(\vec{k}_\parallel) = e^{il\varphi}\,\phi(k_\parallel)\,. \quad (11)$$

For s excitons, the angular-momentum quantum number l takes the values $l = 0, 1$ for LH and $l = -1, 2$ for HH excitons.[3,21] Matrix elements of the exchange interaction between different exciton states (which can be uniquely labeled by l and σ) are given by

$$X^{l\sigma}_{l'\sigma'} = \sum_{\vec{k}_\parallel \vec{k}'_\parallel} e^{-il\varphi}\,\phi^{l\sigma}(k_\parallel)\, V^X_{\lambda_e\lambda_h\lambda'_e\lambda'_h}\, e^{-il'\varphi'}\,\phi^{l'\sigma'}(k'_\parallel). \quad (12)$$

Due to the axial symmetry for a given value of l (l') only one M (M') component of the hole subband function contributes to expression (12). We want to mention that this point was not correctly taken into account in Ref. 9.

The exchange matrix (12) has to be diagonalized separately in the LH and HH subspace. In order to be able to identify longitudinal and transverse exciton states, we perform in the two Γ_5 blocks a transformation from the angular-momentum eigenstates to states which transform as x and y, which with respect to the in-plane wave vector $Q_\parallel \parallel \hat{\mathbf{e}}_x$ are identified as L and T excitons, respectively.

Our choice of $\vec{k}_\parallel = \vec{k}_h$ and $\vec{Q}_\parallel = \vec{k}_e - \vec{k}_h = \vec{k}'_e - \vec{k}'_h$ in Sec. II has the advantage that the hole-subband states with their entanglement due to LH-HH coupling do not depend on $\vec{Q}_\parallel$. However, we have to take into account the coupling term $\frac{\hbar^2}{m_e}\vec{k}_\parallel \cdot \vec{Q}_\parallel$ deriving from the parabolic electron subband dispersion. This is done by including this coupling between s and p excitons in the solution of the exciton integral equation and yields the $Q_\parallel$ dependence due to spatial dispersion.

For the exciton energy including exchange we finally obtain

$$\hbar\omega_Z^{\mathrm{LH}} = \hbar\omega_{\mathrm{ex}}^{\mathrm{LH}}(Q_\parallel) + \tfrac{4}{3}\sum_{\nu\nu'} \Phi_\nu^{\mathrm{LH}}\Phi_{\nu'}^{\mathrm{LH}}[(A+N)K_{\nu\nu'} - \tfrac{1}{2}NQ_\parallel I_{\nu\nu'}(Q_\parallel)], \quad (13)$$

$$\hbar\omega_T^{\mathrm{LH}} = \hbar\omega_{\mathrm{ex}}^{\mathrm{LH}}(Q_\parallel) + \tfrac{1}{3}\sum_{\nu\nu'} \Phi_\nu^{\mathrm{LH}}\Phi_{\nu'}^{\mathrm{LH}}\, AK_{\nu\nu'}, \quad (14)$$

$$\hbar\omega_L^{\mathrm{LH}} = \hbar\omega_{\mathrm{ex}}^{\mathrm{LH}}(Q_\parallel) + \tfrac{1}{3}\sum_{\nu\nu'} \Phi_\nu^{\mathrm{LH}}\Phi_{\nu'}^{\mathrm{LH}}[AK_{\nu\nu'} + \tfrac{1}{2}NQ_\parallel I_{\nu\nu'}(Q_\parallel)]\,, \quad (15)$$

where $N = \frac{4\pi\mu^2}{\varepsilon_\infty}$ and $\hbar\omega_{\mathrm{ex}}(Q_\parallel)$ is the sum of the energy gap, confinement, and binding energy as well as the contribution from the center-of-mass dispersion. For the HH L and T excitons the factor 1/3 has to be omitted and the corresponding HH quantities have to be used in Eqs. (14) and (15). No Z mode exists for the HH excitons. In the above expressions $K_{\nu\nu'}$ and $I_{\nu\nu'}(Q_\parallel)$ are defined by

$$K_{\nu\nu'} = \int dz\, \zeta^*(z)\,\zeta(z)\,\psi_\nu(z)\,\psi^*_{\nu'}(z), \quad (16)$$

$$I_{\nu\nu'}(Q_\parallel) = \int dz\, dz'\, \zeta^*(z)\,\psi_\nu(z)\,\zeta(z')\,\psi^*_{\nu'}(z')\, e^{-Q_\parallel|z-z'|}, \quad (17)$$

and (j=LH,HH)

$$\Phi_\nu^j = \frac{\sqrt{\mathcal{A}}}{2\pi} \int_0^\infty dk_\parallel k_\parallel \, \phi^j(k_\parallel) A^j_{\nu\,-l+\frac{1}{2}}(k_\parallel) \, . \qquad (18)$$

As discussed in Sec. II the dipole-forbidden states are not influenced by the exchange interaction. The Fourier transform of $\phi^j(k_\parallel)$ in Eq. (18) describes the relative electron-hole motion of the QW exciton. At this point we want to emphasize that in the presence of LH-HH mixing the exchange interaction is not a contact interaction any more. Indeed, Φ_ν is proportional to $\phi(\vec{\rho}=0)$ only under the assumption of no LH-HH mixing [then $A_{\nu\,-l+\frac{1}{2}}(k_\parallel) \propto \delta_{1\nu}$]. This point was apparently missed in Ref. 10, where the exchange interaction was treated as a contact interaction. The resulting fine-structure splitting was found to be proportional to $|\phi(0)|^2$, a factor which, however, was calculated using a very elaborate model including LH-HH coupling. The factor Φ_ν also appears in the expression for the oscillator strength and is responsible for the nonvanishing oscillator strength of QW excitons with other than s symmetry.[27]

In Fig. 1 we show the exchange contribution for LH exciton states for a 50-Å quantum well. The main features are the vanishing LT splitting for $Q_\parallel \to 0$ ($\Delta E_{\rm LT} \propto Q_\parallel$ for $Q_\parallel L \ll 1$) and the large anisotropy splitting between Z and L,T exciton states at $Q_\parallel = 0$ as already found by Andreani and Bassani.[10] This splitting has been observed recently in two PL experiments.[14,15] For $Q_\parallel L \gg 1$ the LT splitting is enhanced over the corresponding bulk value and the finite TZ split is due to the analytic exchange. The enhancement factor, which is about 2.5 for $L = 50$ Å, is much smaller than expected by the simple estimate mentioned in the Introduction.

The dispersion of the HH excitons is shown for a 50-Å QW in Fig. 2. It can be seen that the effects due to the center-of-mass dispersion are much more pronounced than the fine-structure splittings.

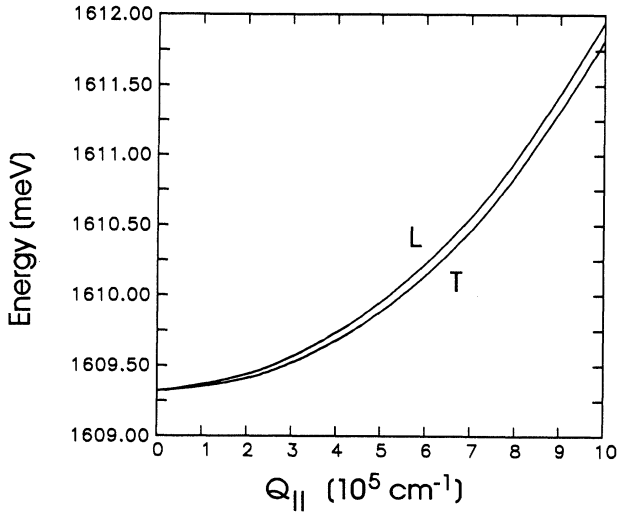

FIG. 2. Energy of the 50-Å HH excitons.

Figure 3 shows similar results for the LH excitons in a 150-Å QW. The strongly nonparabolic exciton dispersion is caused by the corresponding LH subband dispersion.

Figure 4 shows a comparison of our calculated values for the TZ splitting together with experimental results.[14,15] The observed increase of $\Delta E_{\rm TZ}$ with decreasing L is reproduced only qualitatively; however, the experimental results do not yet provide a conclusive picture. The fact that our theoretical values are smaller than those seen in the experiments may be attributed to the neglect of Coulomb coupling to exciton states deriving from higher subbands as well as to continuum states.[5,21]

The LT splitting discussed here should not be confused with the LT splitting in superlattices as studied recently in Ref. 28. The latter is intimately connected with trans-

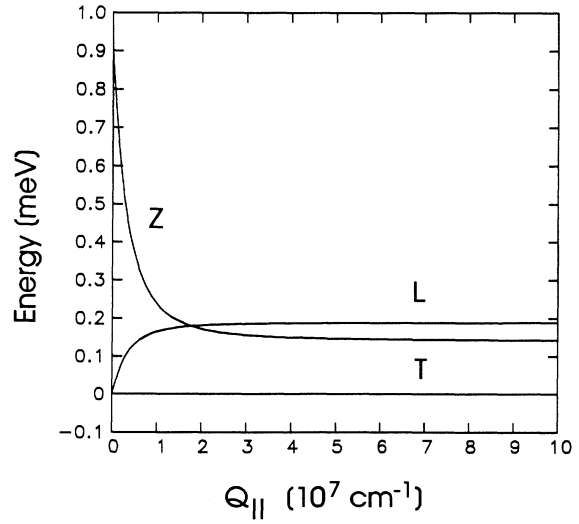

FIG. 1. Exchange contribution for the LH excitons in a 50-Å QW.

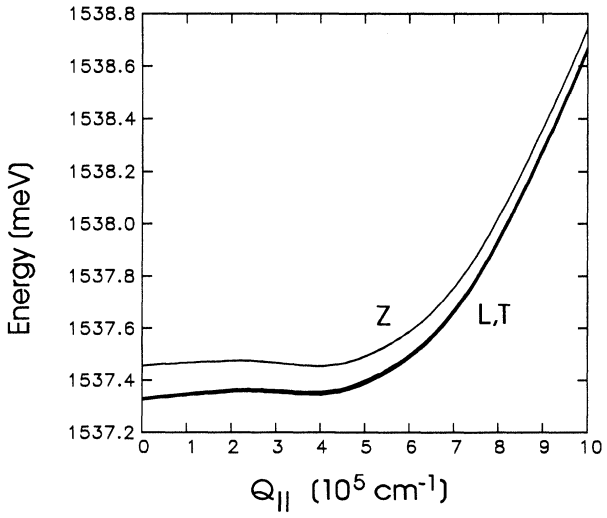

FIG. 3. Energy of the 150-Å LH excitons. The splitting between the L and T exciton is not resolved.

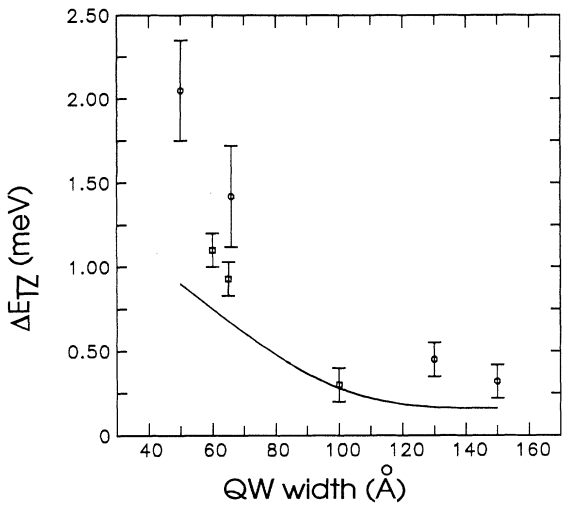

FIG. 4. Theoretical and experimental results from Ref. 15 (○) and Ref. 14 (□) for the TZ splitting for different QW widths.

lational symmetry in the growth direction. Only in this case, for fixed polarization in the xy plane, a transverse mode propagating in the growth direction exists. It is this mode which has been considered for the LT splitting in Ref. 28.

One might expect that the details of the fine structure are more pronounced in QW's with II-VI semiconductors, due to the enhancement of the corresponding bulk LT splits (0.65 meV in CdTe vs 0.08 meV in GaAs). The expression for the bulk LT split reads[24]

$$\Delta E_{\mathrm{LT}}^{\mathrm{3D}} = \tfrac{4}{3}\, N |\phi^{\mathrm{3D}}(0)|^2 \propto N \frac{1}{a_0^3} \tag{19}$$

and the enhanced LT split in CdTe derives from the smaller Bohr radius (by a factor of 2.4), the prefactor N being even smaller in CdTe than in GaAs. In the strict 2D limit the expression for the TZ split is (neglecting the analytic exchange)

$$\Delta E_{\mathrm{TZ}} = \tfrac{4}{3}\, N |\phi^{\mathrm{2D}}(0)|^2 \frac{3}{2L} \propto N \frac{1}{a_0^2}\,, \tag{20}$$

thus, the ratio between the Bohr radii enters only quadratically. This reduces the ratio between the bulk LT splittings in CdTe and GaAs of 8.1 to a corresponding ratio of the TZ splitting in QW's of 3.4. $|\phi(0)|^2$ enters also the expression for the oscillator strength (neglecting LH-HH coupling) and as is well known, the strict 2D limit overestimates this factor so that an even smaller ratio than 3.4 is to be expected. Nevertheless, with the increasing quality of QW's with, e.g., CdTe as well material, an expected TZ splitting of 3 times the values found in GaAs QW's should be detectable in the near future.

IV. THE EXCITON-PHOTON INTERACTION

To derive the exciton-photon interaction we start with the Hamiltonian which describes the interaction of electrons and photons,

$$H_I = \frac{e}{mc} \sum_{\alpha} \int d^3r\, \hat{\psi}_\alpha^\dagger(\mathbf{r})\, \mathbf{p}\cdot\mathbf{A}(\mathbf{r},t)\, \hat{\psi}_\alpha(\mathbf{r}). \tag{21}$$

The field operator $\hat{\psi}_\alpha(\mathbf{r})$ is expanded in terms of annihilation operators $a_{c\sigma\vec{k}_\parallel}$ and $a_{vj\vec{k}_\parallel}$ for electrons in the lowest conduction or highest valence subband, respectively, according to

$$\hat{\psi}_\alpha(\mathbf{r}) = \sqrt{\frac{\nu}{\mathcal{A}}} \sum_{\vec{k}_\parallel} e^{i\vec{k}_\parallel\cdot\vec{\rho}} \left\{ \sum_\sigma a_{c\sigma\vec{k}_\parallel} \zeta(z)\, u^\alpha_{c\sigma\mathbf{0}}(\mathbf{r}) + \sum_{jM} a_{vj\vec{k}_\parallel} \zeta^j_{\vec{k}_\parallel M}(z)\, u^\alpha_{vM\mathbf{0}}(\mathbf{r}) \right\}. \tag{22}$$

At this point the k dependence of the Bloch functions can be neglected. The vector potential is expanded as usual in the eigenmodes of a cube with volume $\mathcal{V}$,

$$\mathbf{A}(\mathbf{r},t) = \sum_{\mathbf{Q}\lambda} \left(\frac{2\pi\hbar v}{Q\mathcal{V}}\right)^{1/2} \hat{\mathbf{e}}_{\mathbf{Q}\lambda} (c_{\mathbf{Q}\lambda} e^{i\mathbf{Q}\cdot\mathbf{r}} + c^\dagger_{\mathbf{Q}\lambda} e^{-i\mathbf{Q}\cdot\mathbf{r}})\,, \tag{23}$$

where $c_{\mathbf{Q}\lambda}$ and $c^\dagger_{\mathbf{Q}\lambda}$ are annihilation and creation operators for photons with 3D wave vector $\mathbf{Q}$ and polarization λ. The in-plane components $\vec{Q}_\parallel$ of $\mathbf{Q}$ are good quantum numbers and with $\vec{Q}_\parallel \parallel \hat{\mathbf{e}}_x$, the two transverse polarization vectors $\hat{\mathbf{e}}_{\mathbf{Q}\lambda}$ may be chosen as $\hat{\mathbf{e}}_{\mathbf{Q}1} = (0,1,0)$ and $\hat{\mathbf{e}}_{\mathbf{Q}2} = (Q_z, 0, -Q_\parallel)/Q$. $v = c/\sqrt{\varepsilon_\infty}$ is the light velocity in the medium with dielectric constant ε_∞. We now introduce hole operators defined by $\beta^\dagger_{-\vec{k}_\parallel} = a_{v\vec{k}_\parallel}$ and rename the electron operators by $\alpha^\dagger_{\vec{k}_\parallel} = a^\dagger_{c\vec{k}_\parallel}$, in order to keep the notation symmetric. These two operators have to be paired to form creation and annihilation operators for excitons with center-of-mass wave vector $\vec{Q}_\parallel$ defined by

$$B^\dagger_{\kappa\vec{Q}_\parallel} = \sum_{\vec{k}_\parallel} \phi^\kappa(\vec{k}_\parallel)\, \alpha^\dagger_{\vec{k}_\parallel + \vec{Q}_\parallel} \beta^\dagger_{-\vec{k}_\parallel}\,, \tag{24}$$

with a similar expression for $B_{\kappa\vec{Q}_\parallel}$. Here κ specifies internal degrees of freedom of the QW exciton. By using the completeness of the $\phi^\kappa(k_\parallel)$ relation (24) can be inverted and used to formulate H_I as

$$H_I = \sum_{\mathbf{Q},\lambda} A^\lambda_{\vec{Q}_\parallel Q_z}(c_{\mathbf{Q}\lambda} + c^\dagger_{-\mathbf{Q}\lambda})(B^\dagger_{\vec{Q}_\parallel} + B_{-\vec{Q}_\parallel}) , \quad (25)$$

with the coupling constant

$$A^\lambda_{\vec{Q}_\parallel Q_z} = \frac{e}{mc}\left(\frac{2\pi\hbar v}{Q\mathcal{L}}\right)^{1/2} \sum_\nu \Phi_\nu \langle c\sigma|\hat{\mathbf{e}}_{\mathbf{Q}\lambda}\cdot\mathbf{p}|vl-\tfrac{1}{2}\rangle \int dz\, \zeta(z)\,\psi_\nu(z)\, e^{iQ_z z} . \quad (26)$$

Exploiting the axial symmetry we have used $l = M + \frac{1}{2}$ (for s excitons), where l is the angular momentum of the envelope function. Care has to be taken when evaluating the matrix element $\langle c\sigma|\hat{\mathbf{e}}_{\mathbf{Q}\lambda}\cdot\mathbf{p}|vl-\frac{1}{2}\rangle$, which gives $1/\sqrt{3}\, p_{cv}$ for T excitons, $1/\sqrt{3}\, Q_z/Q\, p_{cv}$ for L excitons, and $2/\sqrt{3}\ Q_\parallel/Q\ p_{cv}$ for Z excitons. As could be expected, the $\lambda = 1(2)$ mode, which is connected with a TE (TM) field, couples only to T-(L, Z) excitons. Equation (25), due to the inclusion of the finite extension of the wave functions in the z direction and LH-HH mixing represents an extension of Hanamura's formulation.[29]

V. DISPERSION OF POLARITONS

The total Hamiltonian of the exciton-photon system can now be written

$$H = \sum_{\vec{Q}_\parallel} \hbar\omega_{Q_\parallel} B^\dagger_{\vec{Q}_\parallel} B_{\vec{Q}_\parallel} + \sum_{\mathbf{Q},\lambda} \hbar v Q\, c^\dagger_{\mathbf{Q}\lambda} c_{\mathbf{Q}\lambda} + H_I . \quad (27)$$

Heisenberg's equations of motion for the four exciton and photon operators yield for time-periodic solutions and neglecting the antiresonant term dispersion relations of the form

$$\hbar(\omega - \omega_{Q_\parallel}) - \Sigma_{Q_\parallel}(\omega) = 0 , \quad (28)$$

with the self-energy $\Sigma_{Q_\parallel}(\omega)$ given by

$$\Sigma_{Q_\parallel}(\omega) = \frac{1}{\hbar}\sum_{Q_z,\lambda} \frac{2vQ|A^\lambda_{\vec{Q}_\parallel Q_z}|^2}{(\omega + i\delta)^2 - v^2(Q_\parallel^2 + Q_z^2)} . \quad (29)$$

Equation (29) has to be evaluated separately for resonant ($\omega > vQ_\parallel$) and localized ($\omega < vQ_\parallel$) L, T, and Z (the latter being allowed only for LH) polaritons. For the localized L polariton we find a dispersion relation given by

$$\hbar\omega = \hbar\omega_{Q_\parallel} + \frac{2\pi}{\varepsilon_\infty} g\mu^2 \sum_{\nu\nu'} \Phi_\nu\Phi_{\nu'}\left[\alpha I_{\nu\nu'}(\alpha) - Q_\parallel I_{\nu\nu'}(Q_\parallel)\right] . \quad (30)$$

Here $\alpha = \sqrt{Q_\parallel^2 - \omega^2/v^2}$ and the integral $I_{\nu\nu'}(\alpha)$ is already known from the calculation of the nonanalytic exchange interaction [Eq. (17)]. g equals 1/3 (1) for LH (HH). In order to compare with the dispersion relations obtained in Refs. 10 and 16 we want to point out that in our approach the exciton energy $\hbar\omega_{Q_\parallel}$ includes the contribution of both analytical and nonanalytical exchange. This can be seen most easily by the fact that H_I vanishes in the limit $c \to \infty$. This is in contrast to the approach of Refs. 10 and 16 where it was shown that solving the electrostatic Maxwell equations is equivalent to a microscopic calculation of the nonanalytical exchange interaction. Therefore this contribution must not be included in the exciton energy in the approach of Refs. 10 and 16. Substituting the explicit expression for the exchange contribution of the L exciton given in Eq. (15) into Eq. (30) the nonanalytical exchange contribution cancels exactly and we find the dispersion relation

$$\hbar\omega = \hbar\tilde{\omega}_{Q_\parallel} + \frac{2\pi\alpha}{\varepsilon_\infty} g\mu^2 \sum_{\nu\nu'} \Phi_\nu\Phi_{\nu'} I_{\nu\nu'}(\alpha) , \quad (31)$$

where now $\hbar\tilde{\omega}_{Q_\parallel}$ includes only the analytical exchange. For the T and Z polaritons we find in analogy

$$\hbar\omega = \hbar\tilde{\omega}_{Q_\parallel} - \frac{2\pi\omega^2}{\alpha c^2} g\mu^2 \sum_{\nu\nu'} \Phi_\nu\Phi_{\nu'} I_{\nu\nu'}(\alpha) \quad (32)$$

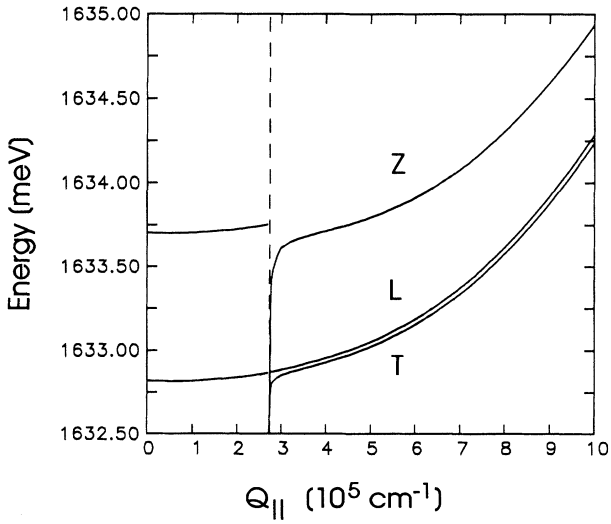

FIG. 5. LH polariton dispersion for a 50-Å QW. Resonant and localized polaritons are separated by the dashed line, which is given by $\hbar\omega = \hbar c Q_\parallel/\sqrt{\varepsilon_\infty}$.

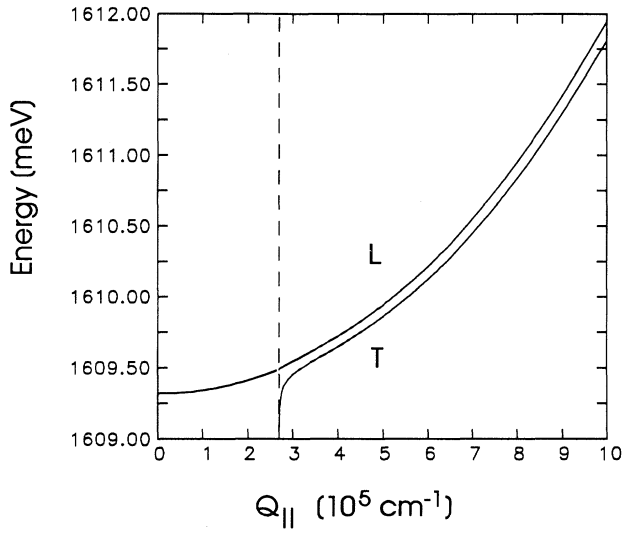

FIG. 6. HH polariton dispersion for a 50-Å QW.

and

$$\hbar\omega = \hbar\tilde{\omega}_{Q_\parallel} - \frac{8\pi}{3\varepsilon_\infty}\mu^2 \sum_{\nu\nu'} \Phi_\nu \Phi_{\nu'} \left\{ \frac{Q_\parallel^2}{\alpha} I_{\nu\nu'}(\alpha) - 2K_{\nu\nu'} \right\} . \tag{33}$$

To obtain the dispersion relations for the resonant polaritons, $\beta = \sqrt{\omega^2/v^2 - Q_\parallel^2}$ must be substituted for α and the exponential function in $I_{\nu\nu'}(\alpha)$ must be replaced by the sine function. Also, for resonant T and Z polaritons the term involving $I_{\nu\nu'}(\alpha)$ has to be multiplied by an extra factor of (-1). The dispersion relations, apart from the differences discussed at the end of Sec. III, are identical to those found in Refs. 10 and 16.

Figures 5 and 6 show the dispersion relations for LH and HH polaritons in a 50-Å $Al_{0.3}Ga_{0.7}As$-GaAs QW. The main features are a large discontinuity of the T and Z modes at the photon line, the L mode being continuous. In the limit $c \to \infty$ the resonant polaritons disappear and the localized ones reduce to the corresponding excitons, i.e., Fig. 6 becomes identical to Fig. 2.

Experiments concerning the QW polaritons have been carried out by Ogawa, Katsuyama, and Nakamura,[13] who made time-of-flight measurements and by Kohl *et al.*,[30] who used a grating coupler to excite the localized polaritons. Both experiments, however, are not sensitive enough to allow for a quantitative verification of the polariton dispersion.

VI. LIFETIMES OF RESONANT POLARITONS

The lifetime τ due to radiative decay of the resonant polaritons is related to the imaginary part Γ of the self-energy (29) by $\tau = \hbar/\Gamma$. The calculation is straightforward and leads to an identical expression for Γ for L and T polaritons at $Q_\parallel = 0$, which agrees with the results obtained in Ref. 31. The corresponding values for τ (in ps) for QW widths of 50, 100, and 150 Å are summarized in Table 1.

TABLE I. Lifetimes of resonant polaritons in picoseconds.

	50 Å	100 Å	150 Å
LH	44.5	102.8	170.5
HH	16.3	21.9	28.7

In a recent letter Deveaud *et al.*[32] reported on the observation of a HH radiative decay width of 10 ± 4 ps in a 45-Å QW of highest quality and at 2 K. They discard the explanation in terms of localized excitons due to impurities and interface defects. Taking into account that our calculation is for a slightly broader well, their result agrees fairly well with our value.

VII. CONCLUSIONS

In conclusion we have calculated the fine structure of QW excitons, which microscopically derives from the electron-hole exchange interaction with parameters known from bulk material. We find that the splitting between longitudinal and transverse QW excitons (with respect to the in-plane exciton wave vector $Q_\parallel$) vanishes linearly for $Q_\parallel \to 0$ and is enhanced over the bulk value for $Q_\parallel \to \infty$. The most important effect of the exchange interaction is the energy shift of the Z exciton relative to the L, T excitons, which is a consequence of the reduced symmetry and was expected from group-theoretical considerations. This splitting has been detected recently in photoluminescence experiments.[14,15] We derived the Hamiltonian which describes in second quantization the interaction of photons with excitons, solved the corresponding equations of motion, and showed that the dispersion relations for localized and resonant polaritons coincide with those obtained by solving Maxwell's equations. For resonant polaritons we calculated a finite lifetime due to decay into electromagnetic modes, which depends on the width of the QW. It is significantly smaller for HH than for LH polariton states. These results are in reasonable agreement with the few existing experimental data[32] but contain more detailed information which we hope will stimulate experiments on this subject.

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