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Rapid Communications

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Exchange interaction in type-II quantum wells

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The heavy-hole-exciton splitting in type-II quantum wells due to the electron-hole exchange interaction is calculated in an effective-mass approximation. Good agreement with the trends observed in recent optically detected magnetic resonance experiments by van Kesteren and colleagues has been found. By comparison of theory with experiment the X-Γ electron-hole exchange interaction for bulk GaAs is determined to be $\simeq$80 meV/molecule.

In spite of prolonged interest in the electron-hole exchange interaction in bulk semiconductors,[1] its order of magnitude is not even known for such a well-investigated compound as GaAs.[2] In type-I GaAs/$Al_xGa_{1-x}As$ quantum wells (QW's) a doublet in the luminescence of the order of 1 meV has been interpreted in terms of an exchange-induced splitting.[3,4] van Kesteren and coworkers[5,6] recently determined the exchange splittings in GaAs/AlAs type-II multiple QW's by optically detected magnetic resonance (ODMR) experiments. In this Rapid Communication, we (i) show that the dependence of the exchange splittings on sample geometry as observed by van Kesteren and colleagues is explained by effective mass theory and (ii) estimate the bulk exchange integral of the indirect X-Γ transition for GaAs by comparison of theory and experiment. To this end we extend the approach of Chen, Gil, Lefebvre, and Mathieu[4] and present for the first time an accurate treatment of the exciton problem in type-II QW's with finite barrier heights.

The effective mass theory for Wannier excitons in semiconductors is readily extended to include the exchange interaction.[1] Neglecting the difference between the Bloch functions in GaAs and AlAs, the exchange interaction in QW's can be expressed in terms of the coordinates of electron and hole $\mathbf{r}_e$ and $\mathbf{r}_h$ by the operator[1,4]

$$\mathcal{H}_{\rm exc} = \Omega \underline{E} \delta(\mathbf{r}_e - \mathbf{r}_h) , \qquad (1)$$

where Ω is the volume of the primitive unit cell and $\underline{E}$ is a 8×8 matrix in the basis of the exciton states constructed from the $S=\frac{1}{2}$ conduction band and $J=\frac{3}{2}$ valence band. Dirac's δ function represents a short-range repulsion between electrons and holes. In a type-II QW recombination takes place between electrons at the X point in AlAs and holes in GaAs at the Γ point. It is clear from Eq. (1) that the separation of electron and hole into different layers leads to a decreased exchange effect. Note that the matrix $\underline{E}$ is not necessarily the same for type-I and type-II QW's since different Bloch states of the electron are involved. The exchange interaction is small compared to the subband energy splittings, the exciton binding energy, and even the Zeeman splittings for the weak fields ($\simeq$0.3 T) at which the ODMR experiments have been carried out.[5,6] We therefore employ perturbation theory and disregard the nondiagonal elements in Eq. (1) (the latter approximation does not hold for a wide QW at high magnetic fields, where heavy- and light-hole excitons may cross[7,8]). The exchange effect on the heavy-hole ground-state excitons considered here is thus the zero-field splitting between the optically allowed and forbidden doublets.

To our knowledge, the only theory of excitons in type-II QW's is due to Duggan and Ralph[9] who investigated the exciton binding energy for a model of impenetrable potentials barriers. In our case, it is essential to allow for finite potential barriers, however, because otherwise the exchange interaction vanishes identically. The anisotropic effective masses of electrons and holes and other parameters are listed in Table I. According to Duggan and coworker[9,10] the mixing of heavy and light holes in the QW structure is accounted for by an enhanced in-plane heavy-hole mass. An isotropic dielectric constant of 11.3 is employed which is the average of the dielectric con-

TABLE I. Effective mass parameters; potential barrier conduction-band X point: 198 meV, potential barrier valence-band Γ point: 526 meV.

	AlAs	GaAs
$m_{e\perp}$	1.1	1.3
$m_{hh\perp}$	0.75	0.34
$m_{e\parallel}$	0.19	0.19
$m_{hh\parallel}$	0.30	0.18
ϵ	11.3	11.3

stants of AlAs (10.9) and GaAs (12.5). The effect of the neglected image potential is discussed later on. Zero-phonon photoluminescence indicates that the electron wave function in the GaAs layer is not purely X-like but contains a component associated to the Γ valley.[11,12] There is experimental[12] and theoretical[13] evidence that the intervalley scattering due to the interface is small (except at narrow-subband resonances), however. As long as the exchange integrals for electrons at the X and Γ points are not orders of magnitude different, this effect can be disregarded for the present problem. We assume localization of the electron and hole at a common interface, i.e., the translational symmetry of a superlattice is disregarded. This approach is justified by experimental evidence of exciton localization[11,12] and will be commented on later.

The exciton envelope function is calculated by solving the generalized eigenvalue problem obtained by an expansion into a sufficiently large nonorthogonal basis. Our expansion reads

$$\Psi_{ex}(\rho,z_e z_h)=\zeta_0^c(z_e)\zeta_0^v(z_h)\sum_{p,q,r} c_{p,q,r}\exp(-|z_e-d_{AlAs}/2|/a_p)\exp(-|z_h+d_{GaAs}/2|/b_q)\exp(-\rho/c_r)\,, \tag{2}$$

where ζ_0^c, ζ_0^v are the ground-state subband envelope functions of electron and heavy hole, ρ is the in-plane distance between both particles, z_e and z_h are the coordinates of electron and hole normal to the interface, d_{GaAs} and d_{AlAs} denote the layer thicknesses, and a_p, b_q, and c_r are length parameters. Hamiltonian matrix elements are calculated as described in Ref. 14. Convergence is found for about 40 basis functions with widely different exponents.

The binding energy of the ground-state excitons in a type-II QW is shown in Fig. 1 as a function of layer thicknesses. The finite overlap of electron and hole envelope functions leads to an increased binding energy of about 10% compared to the infinite potential barrier model.[9] QW's with GaAs layer thicknesses $\gtrsim 30$ Å have a direct gap and are therefore not considered here. The exchange splitting is in first order proportional to the expectation value of Dirac's δ function [Eq. (1)]. The bulk exchange integral is determined by fitting to the experimental data of van Kesteren and co-workers.[5,6] Since the present theory is believed to be more reliable for wide QW's the sample with the largest well width is used, which yields a value of about 80 meV/molecule. Figure 2 displays the nearly exponential dependence of the exchange splittings on the sample geometries. The trends in the theory agree fairly well with experiment, which means that the splittings are indeed proportional to the electron-hole overlap. A deviation between theory and experiments is found, however, which increases systematically when the layer thickness is decreased. Therefore, in the following, we discuss the limitations of the present model.

The theoretical exchange energies are quite sensitive to the choice of the in-plane heavy-hole mass. Its reduction from 0.18 to 0.15, for example, reduces the exchange en-

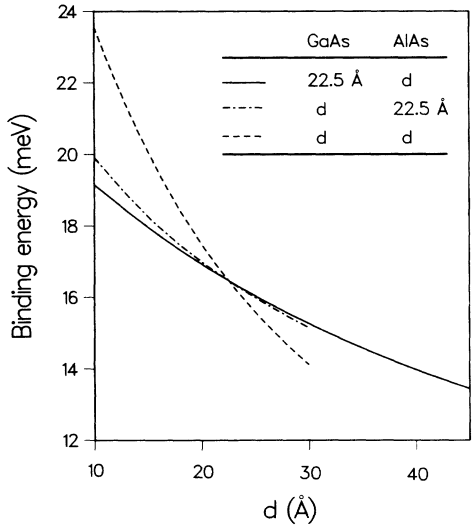

FIG. 1. Theoretical heavy-hole-exciton binding energies in type-II QW's plotted vs thickness parameters. The three curves refer to results obtained by varying (i) the thickness of the AlAs layers for a constant GaAs layer thickness of 22.5 Å, (ii) the thickness of the GaAs layers keeping the AlAs layer at 22.5 Å, and (iii) both layer thicknesses.

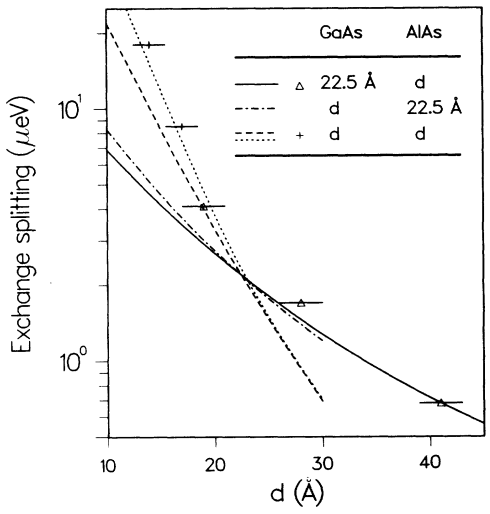

FIG. 2. Theoretical exchange splittings of heavy-hole-exciton transitions in type-II quantum wells. The curves correspond to sample geometries as in Fig. 1. The experimental results of van Kesteren and colleagues[5,6] are represented by triangles (GaAs thickness $\simeq 22.5$ Å) and by crosses (GaAs thickness equals AlAs thickness). The uncertainty in the layer thicknesses is indicated by horizontal lines. The experimental error in the energies is equal or smaller than the size of the markers. The bulk exchange integral is treated as a free parameter and is fitted to the experiment on the sample with largest well width. The dotted line represents an estimation of the effect of particle tunneling into neighboring layers.

ergy by 30%. This reduction is on the other hand almost independent of well width and the systematic increase of the deviation when the well width is decreased cannot be accounted for in this way.

So far we have disregarded the effect of the difference in the dielectric constants of AlAs and GaAs. The dominant correction term in the expansion of the field of the electron as felt by the hole can be represented by the Coulomb field of a negative image charge due to the interface of the GaAs layer (confining the hole) which lies opposite to the AlAs layer confining the electron. Its magnitude is $\simeq 0.6\lambda e$ where $\lambda=(\epsilon_{\mathrm{GaAs}}-\epsilon_{\mathrm{AlAs}})/(\epsilon_{\mathrm{GaAs}}+\epsilon_{\mathrm{AlAs}}) \simeq 0.1$. We therefore expect a small increase of the binding energy ($\lesssim 6\%$) which is comparable to the inaccuracies due to other model assumptions. The deviations in the exchange splitting cannot be explained since with decreasing well width the electron-hole overlap in the z direction is reduced by the Coulomb attraction of the image charge.

The thickness of the thinner layers is only several lattice constants, so the applicability of a δ-function exchange Hamiltonian becomes questionable. Using a sampling function with a spatial extent of about a lattice constant yields corrections of only a few percent with a weak dependence on well width. The same holds for a slight smoothing of square-well potentials.

As mentioned before, our calculations are based on the assumption of exciton localization. Within the limit of thin barriers, however, the exciton wave function ideally extends coherently into neighboring wells. We estimate this effect by comparison of the subband overlaps in a localized QW and in a superlattice at $k_z=0$, neglecting the electron-hole Coulomb interaction. As seen in Fig. 3 (dotted line), the failure of the model can be explained partly by the tunneling of electrons and holes through the barriers. It should be kept in mind that the dotted line is a strictly upper bound, however, and probably overestimates the effect considerably. The assumption of coherence of the exciton wave functions over several wells is also questionable since experiments have been interpreted otherwise.[12]

The most serious deficiency of the present model is the parabolic approximation for the X-valley band structure. In narrow wells the subband energies are large and nonparabolicities are expected to be very important. The "camel-back" band structure can be taken into account by the $\mathbf{k}\cdot\mathbf{p}$ method,[13] but we refrain from extending our theory because of the poor knowledge of the parameters of the AlAs X-point band structure.

The X-Γ exchange integral is expected to be smaller than the Γ-Γ exchange integral, but should not differ by orders of magnitude. Using the present value of 80 meV the exchange splitting in type-I quantum wells is estimated to be about 10 μeV, which is much smaller than the values found by Bauer *et al.*[3] and Chen *et al.*[4] A direct calculation of the bulk exchange integrals should be helpful in resolving these questions.

To summarize, the experimental trends in the electron-hole exchange interaction of Wannier excitons in type-II QW's are semiquantitatively explained. The bulk exchange interaction energy between X-point electrons and Γ-point heavy holes has been estimated to be $\simeq 80$ meV/molecule by comparison with experiments by van Kesteren and colleagues.[5,6] The deviations between theory and experiment for very thin wells (<20 Å) are tentatively ascribed to the nonparabolicity of the conduction band at the X point.

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