Exchange splitting of excitonic levels in types I and II superlattices

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We construct a theory that explains the fine structure of excitonic levels in types I and II semiconductor superlattices. The nature of the anisotropic splitting of the excitonic emission doublet discovered in type II GaAs/AlAs(001) superlattices is explained. We show that this splitting is caused by the low symmetry (C_{2v}) of an ideal heterojunction, which allows for the mixing of states of heavy and light holes even when the hole is normally incident on the heterojunction plane. For the excitonic ground state $e1$–$hh1(1s)$ we calculate the dependence of the axisymmetric exchange splitting and anisotropic exchange splitting on the thickness of the GaAs and AlAs layers. The theoretical results are found to agree with the existing experimental data.

1. INTRODUCTION

The GaAs/AlAs(001) superlattice is a convenient model object for studying type I–type II transitions in heterostructures, since in this superlattice (SL), depending on the layer thickness ratio, the bottom of the conduction band is formed mainly either from states of the Γ-minimum in the GaAs layer or from states of the X-minimum in the AlAs layer.[1–4] In both cases the ideal heterostructure has a D_{2d} point symmetry, in which the lower excitonic level $e1X$–$hh1(1s)$, which is fourfold degenerate if we take into account the electron spin ($\pm 1/2$) and the heavy-hole spin ($\pm 1/2$), must split into an emission doublet E and two optically inactive singlets owing to electron–hole exchange interaction. However, experiments have established that for excitons observed in photoluminescence spectra of a type II GaAs/AlAS superlattice, the E doublet splits into two sublevels which are dipole-active in the [110] and [1$\bar{1}$0] directions.[5–7] There is a need to explain not only the nature of the anisotropy in the exchange interaction but also the fairly large size of the energy gap between the sublevels, $\delta=\varepsilon_{[110]}-\varepsilon_{[1\bar{1}0]}$, and the fact that in a single SL with fixed layer thicknesses two classes of excitons exist simultaneously, excitons for which the absolute values of δ coincide and are fixed but whose signs are opposite. Finally, it must be understood why the exchange-interaction anisotropy is observed in a type II SL but not in a type I. In Ref. 8 an assumption was made that the anisotropy is related to the lower symmetry C_{2v} of the ideal GaAs/AlAs(001), which allows for mixing of states of heavy and light holes (l–h mixing) even in normal incidence of a hole on the interface. In this paper we develop a consistent theory of the exchange interaction of an electron and hole in an exciton for GaAs/AlAs superlattices of both types that allows for l–h mixing at a heterojunction. We calculate the dependence of isotropic and anisotropic exchange splitting on the thickness of GaAs and AlAs layers and compare the results with the available experimental data.

2. THE EXCITON WAVE FUNCTION

In this section we give the expression for the wave function of an exciton in a superlattice that is a sandwich of GaAs and AlAs layers of thickness a and b, respectively. The range of a and b considered is such that the energy gap between the lower minibands of a Γ- and X-electron (the $e1\Gamma$ and $e1X$ minibands) is large compared to the size of the Γ–X interaction, so that the SL can be characterized by a certain type. The interface, where the Γ and X states are strongly mixed, has been analyzed in Ref. 9.

Since tunneling of a heavy hole between GaAs layers is negligible, in setting up the excitonic states we can assume that the hole in each exciton is in one of these layers. This makes it possible in a variational calculation to represent the trial wave function of a free exciton in the form of the product

$$\Psi^{\rm exc}=S^{-1/2}\,\Psi_e(z_e,\rho)\psi_{hh1}(z_h), \tag{1}$$

where S is the sample's area in the interface plane, $z_{e,h}$ is the coordinate of an electron or hole, $\psi_{hh1}(z_h)$ is the wave function of a hole at the bottom of the lower hole $hh1$ subband in AlAs/GaAs/AlAs, a structure with a single quantum well with a width a, and $\boldsymbol{\rho}$ is a two-dimensional vector with coordinates x_e-x_h and y_e-y_h.

In a type I superlattice, in which the width of the lower electron $e1\Gamma$ miniband exceeds the exciton binding energy, we can employ the following approximation for $\Psi_e(z_e)$:[10]

$$\Psi_e(z_e,\rho)=F_{3D}(z_e,\rho)w_{e1\Gamma}(z_e)\,|\Gamma_1,s\rangle, \tag{2}$$

where $|\Gamma_1,s\rangle$ is the Bloch function of an electron with spin $s=\pm 1/2$ at the Γ-point, $w_{e1\Gamma}(z_e)$ the envelope of the wave function of an electron in the superlattice calculated in the Kronig–Penney model, the wave function of relative motion is

$$F_{3D}(z_e,\rho)=(\pi a_\parallel\, a_\perp^2\,)^{-1/2}$$
$$\times\exp\{-[(z_e/a_\parallel\,)^2+(\rho/a_\perp\,)^2]^{1/2}\}, \tag{3}$$

where $a_\perp$ and $a_\parallel$ are the variational parameters, and z_e is measured from the center of the well with the hole.

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In the tight-binding approximation for a heavy hole, the wave function of a free-exciton state with a given projection k_z of the wave vector on the z axis is

$$\Psi_{k_z}^{\text{exc}}=(NS)^{-1/2}N\sum_m \exp(ik_z md)\Psi_e \times(z_e-md)\psi_{hh1}(z_h-md),$$

where N is the number of periods in the superlattice, m is the number of the GaAs layer, and $d=a+b$ is the SL period.

Because of the large longitudinal effective mass of an X-electron, in type II superlattices the tight-binding approximation can be used for both the electron and the hole:

$$\Psi_{k_z}^{\text{exc}}=\frac{1}{\sqrt{N}}\sum_m \exp\{ik_z md\}(C_+|m,+\rangle+C_-|m,-\rangle). \tag{4}$$

Here $|m,\pm\rangle$ is the state of a quasi-two-dimensional exciton with a hole in the mth layer and an electron in the neighboring left or right AlAs layer:

$$|m,\pm\rangle=S^{-1/2}\Psi_e^{\pm}(z_e-md,\rho)\psi_{hh1}(z_h-md), \tag{5}$$

$$\Psi_e^{\pm}(z_e,\rho)=F_{2D}(\rho)\left[u_0\left(z_e\pm\frac{d}{2}\right)|X_1\rangle+v_0\left(z_e\pm\frac{d}{2}\right)|X_3\rangle\right], \tag{6}$$

where

$$F_{2D}(\rho)=(2/\pi a_\perp^2)^{1/2}\exp(-\rho/a_\perp) \tag{7}$$

is the relative-motion function.

In Eq. (6) we allowed for the fact that in GaAs and AlAs crystals the conduction band near the X-point consists of two close bands X_1 and X_3 and we introduced the respective Bloch functions $|X_1\rangle$ and $|X_3\rangle$. Hence, instead of one envelope function $w(z)$ for a Γ-electron we had to introduce two envelopes, $u(z)$ and $v(z)$. It is convenient to write the Hamiltonian that acts on these functions as a 2×2 matrix (see, e.g., Ref. 11):

$$\hat{H}_x=E_x^0+\frac{\hbar^2k_z^2}{2m_x^{\parallel}}+\frac{\hbar^2k_\perp^2}{2m_x^{\perp}}+\begin{pmatrix}\Delta/2 & -iRk_z\\ iRk_z & -\Delta/2\end{pmatrix}, \tag{8}$$

where $\hat{k}_\alpha=-i\partial/\partial r_\alpha$ ($\alpha=x,y,z$), $E_x^0(z)$ is the energetic position of the center between the X_1 and X_3 bands, which changes abruptly as the interface is passed, and Δ the gap between the bands; the constant R describes the **kp** interaction between these two bands responsible for the formation of the double-humped structure of the conduction band X_1 in the bulk of the material. For simplicity we ignore the difference in the values of $m_x^{\parallel}$, $m_x^{\perp}$, Δ, and R in neighboring layers. The superscript 0 in (6) signifies that the functions u and v are calculated for a structure consisting of an AlAs layer (with its center at point $\pm d/2$) and adjoining semi-infinite barrier GaAs layers. For estimates we can put the parameter $a_\perp$ in (7) equal to the Bohr radius of the two-dimensional exciton,

$$a_{2D}=\hbar\kappa/2e^2\mu_\perp,$$

where κ is the dielectric constant, which screens the electron–hole Coulomb interaction, and $\mu_\perp$ is the reduced mass equal to $m_{hh1}^{\perp}m_x^{\perp}/(m_{hh1}^{\perp}+m_x^{\perp})$, with the effective mass $m_{hh1}^{\perp}$ describing the motion of an $hh1$ hole in the interface plane.

With the symmetry of the superlattice taken into account, the coefficients $C_\pm$ in (4) are linked either by the relation $C_-=C_+^*$ or by the relation $C_-=-C_+^*$. Then, normalizing $\Psi_{k_z}^{\text{exc}}$ to unity yields $|C_\pm|=1/\sqrt{2}$. Note that if we allow for the overlap of the wave functions of an X-electron in neighboring AlAs layers, the exciton level splits into sublevels with $C_+=C_-$ and $C_+=-C_-$. If we ignore this overlap and instead allow for the overlap of the wave functions of holes in neighboring GaAs layers, the split electronic states are described by the function (4) with $C_+=\exp\{ik_zd/2\}$ and $C_-=\pm\exp\{-ik_zd/2\}$.

Low-temperature luminescence usually involves excitons localized at the imperfections of the heterostructures, say, on fluctuations of layer widths, or excitons bound by impurity centers, if such centers exist. When the characteristic localization radius L exceeds the Bohr radius $\alpha_\perp$, the localized-exciton wave function is obtained from (5) simply by replacing the normalization factor $S^{-1/2}$ with a smooth envelope $f(\mathbf{R}_\perp)$, where the vector $\mathbf{R}_\perp$ specifies the position of the exciton center of mass in the layer plane, that is,

$$\Psi_{\text{loc},\pm}^{\text{exc}}=f(\mathbf{R}_\perp)\Psi_e^{\pm}(z_e-md,\rho)\psi_{hh1}(z_h-md). \tag{9}$$

In reality the potential localizing the exciton is asymmetric with respect to the center of a GaAs or AlAs layer, and the asymmetry of the respective potential energy exceeds the overlap integral, which determines the width of the X miniband or the miniband $hh1$ of an ideal SL. For this reason, an electron in each localized exciton is definitely either to the right or left of the layer in which a hole is excited.

3. SIZE QUANTIZATION OF AN X-ELECTRON

Let us calculate the envelopes $u_0(z)$ and $v_0(z)$ of the wave function of an X-electron in the GaAs/AlAs/GaAs structure. We place the origin of the z axis at the center of the AlAs layer. Then in the ground state $e1X$ the functions u_0 and v_0 are respectively even and odd under the $z\to-z$ transformation:

$$u_0(z)=D_1\cos(k_1z)+D_2\cos(k_2z),$$
$$v_0(z)=D_1\theta_1\sin(k_1z)+D_2\theta_2\sin(k_2z) \tag{10a}$$

inside the AlAs layer, and

$$u_0(z)=G_1\exp\{-\kappa_1\tilde{z}\}+G_2\exp\{-\kappa_2\tilde{z}\},$$
$$v_0(z)=(G_1\nu_1\exp\{-\kappa_1\tilde{z}\}+G_2\nu_2\exp\{-\kappa_2\tilde{z}\})\,\text{sgn}(z) \tag{10b}$$

outside the AlAs layer. Here $\tilde{z}=(|z|-b/2)$, k_1^2 and k_2^2 are the two roots of the dispersion equation

$$\left(\varepsilon_x-\frac{\hbar^2k^2}{2m_x^{\parallel}}\right)^2=\left(\frac{\Delta}{2}\right)^2+R^2k^2 \tag{11a}$$

for an X-electron in the interior of AlAs, and $i\kappa_1$ and $i\kappa_2$ satisfy a similar equation,

$$\left(\varepsilon_x-\bar{\Delta}+\frac{\hbar^2\kappa^2}{2m_x^{\parallel}}\right)^2=\left(\frac{\Delta}{2}\right)^2-R^2\kappa^2, \tag{11b}$$

in the interior of GaAs, with $\bar{\Delta}=E_x^0(\mathrm{GaAs})-E_x^0(\mathrm{AlAs})$, and the energy ε_x measured from $E_x^0(\mathrm{AlAs})$. It is assumed that this energy satisfies the inequalities $\varepsilon_x+\Delta/2<\Delta,\bar{\Delta}$, so that all four roots of Eq. (11b) have finite real parts and $\kappa_2=\kappa_1^*$. The special case of thick AlAs layers, when the size quantization level lies between the minimum and maximum of the double-humped structure of the X_1 band is not considered here. Hence $\varepsilon_x>-\Delta/2$, and of the two roots k_1 and k_2 one is real and the other imaginary. In (10) we have introduced the coefficients $\theta_{1,2}$ and $\nu_{1,2}$ defined as follows:

$$\theta=\frac{Rk}{\varepsilon_x-\Delta/2-\hbar^2k^2/2m_x^{\parallel}},\quad \nu=\frac{R\kappa}{\varepsilon_x-\Delta/2-\bar{\Delta}+\hbar^2\kappa^2/2m_x^{\parallel}}. \tag{12}$$

When the masses $m_x^{\parallel}$ in the neighboring layers are the same, the boundary conditions reduce to the requirement that the functions u_0 and v_0 and their derivatives du_0/dz and dv_0/dz be continuous at interfaces. Substituting (10) into these boundary conditions and solving the resulting system of four homogeneous linear equations, we arrive at an equation for the energy of states with an even function $u_0(z)$:

$$\begin{aligned}&(\theta_1 s_1-Tk_1s_1-Pc_1)(\theta_2k_2c_2-T|\kappa_1|^2c_2+Q\,k_2\,s_2)\\&\quad=(\theta_2s_2-Tk_2s_2-Pc_2)(\theta_1k_1c_1-T|\kappa_1|^2c_1+Q\,k_1\,s_1).\end{aligned} \tag{13}$$

Here $c_j=\cos(k_jb/2)$, $s_j=\sin(k_jb/2)$,

$$P=\frac{\nu_1\kappa_2-\nu_2\kappa_1}{\kappa_2-\kappa_1},\quad Q=\frac{\nu_2\kappa_2-\nu_1\kappa_1}{\kappa_2-\kappa_1},\quad T=\frac{Q-P}{\kappa_2+\kappa_1}.$$

A similar transcendental equation for solutions with the odd function $u_0(z)$ can be found from (13) by simply replacing c_j with s_j and s_j with $-c_j$.

4. SIZE QUANTIZATION OF A HOLE

We employ the Bloch function basis of the representation Γ_8 of the T_d group in the form

$$\begin{aligned}|3/2\rangle&=-\uparrow(X+iY)/\sqrt{2},\quad |1/2\rangle\\&=[2\uparrow Z-\downarrow(X+iY)]/\sqrt{6},\\|-1/2\rangle&=[2\downarrow Z+\uparrow(X-iY)]/\sqrt{6},\quad |-3/2\rangle\\&=-\downarrow(X-iY)/\sqrt{2},\end{aligned} \tag{14}$$

where $\uparrow$ and $\downarrow$ are spin columns, and X, Y, and Z are the coordinate functions of representation Γ_{15} of the T_d group. We expand the wave function ψ_{hh1} in the basis functions (14):

$$\psi_{hh1}=\sum_j\varphi_j(z)\,|j\rangle, \tag{15}$$

where $j=\pm3/2,\pm1/2$. If we allow for l–h mixing, the simplest boundary conditions for the envelopes allowed by the C_{2v} symmetry and time symmetry and that retain the continuity of net particle flux through the boundary are[8]

$$\varphi_j^A=\varphi_j^B,\quad \nabla_h^j\varphi_j^A=\nabla_h^j\varphi_j^B+\frac{2}{\sqrt{3}}t_{l-h}\sum_{j'}(J_xJ_y)_{jj'}\varphi_{j'}^B. \tag{16}$$

Here the superscripts A and B indicate that the respective quantity belongs to the GaAs and AlAs layers, respectively, t_{l-h} is a real factor,

$$\nabla_h^{\pm3/2}=a_0\frac{m_0}{m_{hh}}\frac{\partial}{\partial z},\quad \nabla_h^{\pm1/2}=a_0\frac{m_0}{m_{lh}}\frac{\partial}{\partial z}, \tag{17}$$

a_0 is the lattice constant (in GaAs $a_0=5.6$ Å), m_0 is the free-electron mass, m_{hh} and m_{lh} are the effective masses of a heavy and light hole (we ignore the difference of hole masses in layers A and B), the J_α are the angular-momentum matrices of states with total angular momentum equal to 3/2, and the factor a_0m_0 was introduced into (17) so that the l–h mixing coefficient t_{l-h} in (16) is a dimensionless quantity. In the basis (14) the only nonzero elements of the matrix

$$(J_xJ_y)\equiv\frac{1}{2}(J_xJ_y+J_yJ_x)=\frac{\sqrt{3}}{2}\begin{pmatrix}0&0&-i&0\\0&0&0&-i\\i&0&0&0\\0&i&0&0\end{pmatrix}$$

are the off-diagonal elements with $|j-j'|=2$, so that the state $|3/2\rangle$ mixes with $|-1/2\rangle$ and the state $|-3/2\rangle$ mixes with $|1/2\rangle$.

Let us examine the origin and properties of the additional term in the boundary conditions (16). At $t_{l-h}=0$ these are simply the standard boundary conditions for the envelopes of the wave function of a heavy or light hole when the hole impinges on the interface at right angles.

An ideal GaAs/AlAs(001) heterojunction is an atomic As plane to the left and right of which Ga and Al layers are positioned so that the Ga–As atom pairs lie in the (110) plane and Al–As pairs lie in the $(1\bar{1}0)$ plane. Hence, a single heterojunction is characterized by the point symmetry C_{2v}, which incorporates, in addition to the identity transformation, the reflection planes $\sigma_v\|$ (110) and $\sigma_v'\|$ $(1\bar{1}0)$ and the twofold-symmetry axis C_2.

In the C_{2v} group the pairs of functions $|3/2\rangle$ and $|-3/2\rangle$, and $|1/2\rangle$ and $|-1/2\rangle$ are transformed via equivalent spinor representations and the function z or xy is an invariant. This implies that the symmetry allows for mixing at the heterojunction of states $|\pm3/2\rangle$ and $|\mp1/2\rangle$, which differs from the well-known l–h mixing emerging from oblique incidence of a hole owing to off-diagonal elements of the Luttinger Hamiltonian, since in normal incidence the Hamiltonian is diagonal.

The additional terms linear in φ_j and $\partial\varphi_j/\partial z$ and responsible for l–h mixing can enter into (16) in both the boundary condition for the envelopes and that for the derivatives. The coefficients of φ_j and $\partial\varphi_j/\partial z$ in these additional terms can be found by doing numerical calculations via the tight-binding approximation or the pseudopotential

method in the manner of Ref. 11, where the coefficients of Γ–X mixing of electronic states in the GaAs/AlAs superlattice were found. According to Ref. 11, the envelopes $w(z)$, $u(z)$, and $v(z)$ of the wave function of an electron in the Γ_1, X_1, and X_3 states, respectively, and the function $(m_x^{\parallel})^{-1}\partial u/\partial z$ are continuous at the heterojunction, and to the boundary conditions for the derivatives $\partial w/\partial z$ and $\partial v/\partial z$ terms are added proportional to the boundary value of v and w, respectively. In describing l–h mixing in this paper we postulate a similar structure of the additional terms in the boundary conditions (16). Symmetry considerations imply that these terms can be represented as the following linear combination:

$$\sum_p t_p \hat{M}_p \varphi_b,$$

where the t_p are constants, the boundary values of the envelopes φ_j are represented in the form of a four-component column φ_b, and the $\hat{M}_p$ ($p=1,2,...$) are linearly independent 4×4 matrices invariant under a $\hat{D}^{-1}(g)\hat{M}_p\hat{D}(g)$ transformation, with the matrix $\hat{D}(g)$ representing the transformation that the basis functions (14) undergo under an operation g belonging to group C_{2v}, a subgroup of T_d. Of the complete set of sixteen 4×4 matrices only the following are invariants in the C_{2v} group: J_z^2, $J_x^2+J_y^2=(15/4)\hat{I}-J_z^2$, $\{J_xJ_y\}$, and $V_z\equiv\{J_z, J_x^2-J_y^2\}$, with $\hat{I}$ the identity matrix. The first two matrices are diagonal and do not lead to mixing of heavy and light holes. Since the $\{J_xJ_y\}$ matrix is symmetric and the V_z matrix is antisymmetric under time inversion,[12] the coefficient $t_3=t_{l-h}$ must be real and the coefficient t_4 imaginary. The particle flux at the boundary is conserved if the quantity

$$i\sum_j(\varphi_j^*\nabla_h^j\varphi_j-\varphi_j\nabla_h^j\varphi_j^*)$$

is continuous, from which it follows that $t_4-t_4^*=2t_4=0$. This completes the justification of the structure of the additional term in the boundary conditions (16) for a single GaAs/AlAs heterojunction. As noted in Sec. 1, the GaAs/AlAs(001) superlattice is characterized by the D_2d symmetry, which includes the mirror rotation S_4 about the center of the GaAs and AlAs layers. Hence, the boundary conditions for the AlAs/GaAs heterojunction can be obtained from (16) as a result of an S_4 transformation in which z is transformed into $-z$ and the function xy [or $\{J_xJ_y\}$] into $-xy$ [or $-\{J_xJ_y\}$]. This implies that the boundary conditions (16) have the same form with the same coefficient t_{l-h} for the GaAs/AlAs and AlAs/GaAs heterojunctions if in both cases the superscripts A and B are referred to the GaAs and AlAs layers, respectively.

The coefficient t_{l-h} is not small in the relativistic limit. Indeed, if we ignore the spin–orbit coupling and spin, the state of the hole at a Γ-point is threefold degenerate and is transformed according to the representation Γ_{15} of the group T_d. As we did in Eq. (14), we denote the three Bloch functions of Γ_{15} by $X(\mathbf{r})$, $Y(\mathbf{r})$, and $Z(\mathbf{r})$. If we now use arguments based on the fact that the symmetry of the heterojunction is low, we can easily show that when a free hole is normally incident on the junction, there must be mixing of the $\exp\{ik_zz\}X(\mathbf{r})$ and $\exp\{ik_zz\}Y(\mathbf{r})$ states or equivalently, the amplitude coefficients of reflection (or transmission) of a hole, $(X+Y)/\sqrt{2}$ and $(X-Y)/\sqrt{2}$, are generally different. Hence, the coefficient t_{l-h} in (16) is proportional to the nonrelativistic coefficients of mixing of the X and Y states at a heterojunction.

Note that incorporating an additional term in (16) is equivalent to allowing for a delta-function contribution to the hole Hamiltonian,

$$H_{l-h}=\frac{2}{\sqrt{3}}(J_xJ_y)\alpha_0U\sum_n\zeta_n\delta(z-z_n), \tag{18}$$

where $U=t_{l-h}\hbar^2/2m_0a_0^2$, z_n is the coordinate of the nth heterojunction, and $\zeta=\pm1$ for a heterojunction of the types BA and AB, respectively. The factor ζ appears in (18) because the boundary conditions (16) retain their form for both types of heterojunction, GaAs/AlAs and AlAs/GaAs, so that, according to (16), the second derivative at the junction z_n incorporates the term

$$\frac{\partial}{\partial z}\nabla_h^j\varphi_j=\pm\frac{2}{\sqrt{3}}t_{l-h}\sum_{j'}(J_xJ_y)_{jj'}\varphi_{j'}^B\delta(z-z_n).$$

The operator (18) balances this term, which occurs in the Schrödinger equation when the operator of kinetic energy of a hole acts on φ_j. In calculating the hole wave function we use the boundary conditions in the form (16). When studying the mixing of two close size-quantization levels of a heavy and light hole, it is convenient to first calculate these levels at $t_{l-h}=0$, and then switch on the interaction between the two holes, H_{l-h}. According to (18) the operator h_{l-h} mixes the envelope functions of different parity in z, in contrast to the parity-conserving hole perturbation operator in an external electric field or in a uniaxially strained sample. The form of (18) graphically illustrates the short-range nature of l–h mixing. Obviously, within the range of applicability of the effective mass method the coefficient t_{l-h} can be assumed constant, independent of the hole energy, inside the GaAs and AlAs layers. This coefficient, however, can depend on the type of composite materials, e.g., on the composition if one of the materials is a solid solution. When the difference between m_{hh} and m_{lh} is large, that is, when the splitting of the subbands of heavy and light holes is much larger than the size of the l–h interaction proportional to t_{l-h}, this interaction mixes free states but cannot localize a hole at an isolated heterojunction. The opposite limiting case of almost identical masses m_{hh} and m_{lh} requires separate consideration and is not examined here.

The pair of wave functions for the hole states at the bottom of the lower subband $hh1$ in a structure with a single GaAs well transforms according to the spinor representation Γ_6, with the spin columns $\uparrow$ and $\downarrow$ transforming according to the same representation. For $t_{l-h}\neq0$ these functions can be written as

$$\varphi_{hh1,\pm3/2}(z_h)=C(z_h)\,|\pm3/2>\pm iS(z_h)\,|\mp1/2>, \tag{19}$$

where $C(z)$ and $S(z)$ are real functions. If we select the origin at the center, we get

$$C(z)=F_h\cos k_h z,\quad S(z)=F_l\sin k_l z \quad \text{if} \quad |z|<a/2,$$

$$C(z)=F_h\cos\phi_h\exp[-\kappa_h(|z|-a/2)] \tag{20}$$

$$S(z)=F_l\sin\phi_l\exp[-\kappa_l(|z|-a/2)]\,\mathrm{sgn}(z) \quad \text{if} \quad |z|>a/2.$$

Here

$$k_h=\sqrt{\frac{2m_{hh}\varepsilon_h}{\hbar^2}},\quad \kappa_h=\sqrt{\frac{2m_{hh}(V_h-\varepsilon_h)}{\hbar^2}},$$
$$k_l=\sqrt{\frac{2m_{lh}\varepsilon_h}{\hbar^2}},\quad \kappa_l=\sqrt{\frac{2m_{lh}(V_h-\varepsilon_h)}{\hbar^2}}, \tag{21}$$

where ε_h is the size quantization energy of the hole, V_h is the size of the discontinuity of the valence band at the heterojunction, $\phi_h=1/2k_h a$, and $\phi_l=1/2k_l a$. The equation for ε_h and the expression for the F_l-to-F_h ratio is

$$a_0^2(k_h\tan\phi_h-\kappa_h)(k_l\cot\phi_l+\kappa_l)+t_{l-h}^2\frac{m_{hh}m_{lh}}{m_0^2}=0, \tag{22}$$

$$\frac{F_l}{F_h}=t_{l-h}\frac{m_{lh}}{m_0 a_0}\frac{\cos\phi_h}{k_l\cos\phi_l+\kappa_l\sin\phi_l}. \tag{23}$$

In the first order in t_{l-h}, the equation for the lower-level energy ε_{hh1} is reduced to the unperturbed equation $k_h\tan\phi_h=\kappa_h$.

5. EXCHANGE INTERACTION IN AN EXCITON

We write the operator of the electron–hole exchange interaction in a bulk A_3B_5 semiconductor (see Ref. 13) in the form

$$V^{\text{exch}}=-\frac{2}{3}a_0^3\delta(r_e-r_h)\left(\varepsilon_0\boldsymbol{\sigma}\mathbf{J}+\varepsilon_1\sum_\alpha\sigma_\alpha J_\alpha^3\right), \tag{24}$$

where the σ_α are the Pauli spin matrices, and $\alpha=x,y,z$ are the principal symmetry axes. Experiments[5,14] have shown that the constant factor ε_1 of the cubic invariant is small compared to the constant factor ε_0 of the spherical invariant $\boldsymbol{\sigma}\mathbf{J}$.

If we do not allow for the exchange interaction, the ground state ($n=1$) of the exciton, $e1\Gamma$–$hh1$, in a type II superlattice is fourfold degenerate. Calculating the matrix elements $V^{\text{exch}}_{s'j',sj}$ of the operator (24) between the excitonic states $|s,j\rangle$ and $|s',j'\rangle$ with given electron and hole spins, within the basis $|-1/2,3/2\rangle$, $|1/2,-3/2\rangle$, $|1/2,3/2\rangle$, and $|-1/2,-3/2\rangle$ we obtain

$$\hat{V}^{\text{exch}}=\frac{1}{2}\begin{pmatrix}\delta_0 & i\delta_2 & 0 & 0\\ -i\delta_2 & \delta_0 & 0 & 0\\ 0 & 0 & -\delta_0 & \delta_1\\ 0 & 0 & \delta_1 & -\delta_0\end{pmatrix}. \tag{25}$$

Here $|\delta_0|$ is the isotropic exchange splitting between states with projections $M=\pm1$ and $M=\pm2$ of the total angular momentum, $|\delta_1|$ the splitting between the optically inactive singlet states

$$(|1/2,\ 3/2\rangle\pm|-1/2,-3/2\rangle)/\sqrt{2},$$

and the parameter δ_2 describes the splitting of the emission doublet into the states

$$(|-1/2,\ 3/2\rangle\pm|1/2,-3/2\rangle)/\sqrt{2},$$

which are optically active in the polarizations $\mathbf{e}\|$ [110] and $\mathbf{e}\|$ [1$\bar{1}$0], respectively, and for which $(\delta_0\mp\delta_2)/2$ are the respective exchange energies.

For a type I superlattice the excitonic wave function has been defined in Eqs. (1), (2), and (19). For the other two parameters, $\delta_0^{(\mathrm{I})}$ and $\delta_1^{(\mathrm{I})}$ it is sufficient to give expressions obtained on the assumption that there is no l–h mixing:

$$\delta_0^{(\mathrm{I})}=\left(\varepsilon_0+\frac{9}{4}\varepsilon_1\right)\frac{2a_0^3}{\pi a_{\|}a_\perp^2}\int C^2(z)e_{e1\Gamma}(z)\,dz, \tag{26a}$$

$$\delta_1^{(\mathrm{I})}=\frac{\varepsilon_1}{\varepsilon_0+9\varepsilon_1/4}\delta_0^{(\mathrm{I})}. \tag{26b}$$

Since $|\varepsilon_1|\ll|\varepsilon_0|$ holds, we also have $|\delta_1^{(\mathrm{I})}|\ll|\delta_0^{(\mathrm{I})}|$. Allowing for the mixing of states of heavy and light holes leads to corrections in (26a) proportional to t_{l-h}^2.

For a free-electron state (4) in an ideal type II superlattice the emission doublet does not split either, that is, $\delta_2^{(\mathrm{II})}=0$, the parameter $\delta_1^{(\mathrm{II})}$ is related to $\delta_0^{(\mathrm{II})}$ through Eq. (26b), and for $\delta_0^{(\mathrm{II})}$ we have the following expression (if we ignore l–h mixing):

$$\delta_0^{(\mathrm{II})}=\left(\varepsilon_0+\frac{9}{4}\varepsilon_1\right)\frac{4a_0^3}{\pi a_\perp^2}\int C^2(z)\phi_e^2\left(z\pm\frac{d}{2}\right)dz, \tag{27}$$

where $\phi_e^2(z)=u_0^2(z)+v_0^2(z)$; the function $C(z)$ has been defined in (20) and the functions u_0 and v_0 in (6). Either of the two signs in the independent variable $z\pm d/2$ can be chosen.

For a localized exciton in a type II superlattice with the electron to the right (an AB exciton) or left (a BA exciton) of the GaAs layer in which the hole resides, the parameter $\delta_2^{(\mathrm{II})}$ is finite and is given by the formula

$$\delta_2^{(\mathrm{II})}=\left(\varepsilon_0+\frac{7}{8}\varepsilon_1\right)\frac{16a_0^3}{\sqrt{3}\,\pi a_\perp^2}\int C(z)S(z)\phi_e^2\left(z\pm\frac{d}{2}\right)dz, \tag{28}$$

and for $\delta_0^{(\mathrm{II})}$ we still have formula (27). The reader will recall that $C(z)$ and $S(z)$ are, respectively an even and odd function of z. Hence, the value of $\delta_0^{(\mathrm{II})}$ is independent of the electron position with respect to the hole, while the AB and BA excitons have $\delta_2^{(\mathrm{II})}$ of opposite sign. Thus, the two types of excitons with opposite signs of the exchange splitting parameter are localized AB and BA excitons. The anisotropy axes of the exchange interaction are fixed by the directions of the reflection planes of the C_{2v} group, that is, by the direction of chemical bonds at the interface. The difference in the signs of the splitting $\varepsilon_{[110]}-\varepsilon_{[\bar{1}10]}$ for these excitons is caused by the fact that under the S_4 transformation the (110) and (1$\bar{1}$0) planes transform into each other.

In deriving (28) we employed the expression (9) for the wave function of a localized exciton valid for $L\gg a_\perp$,

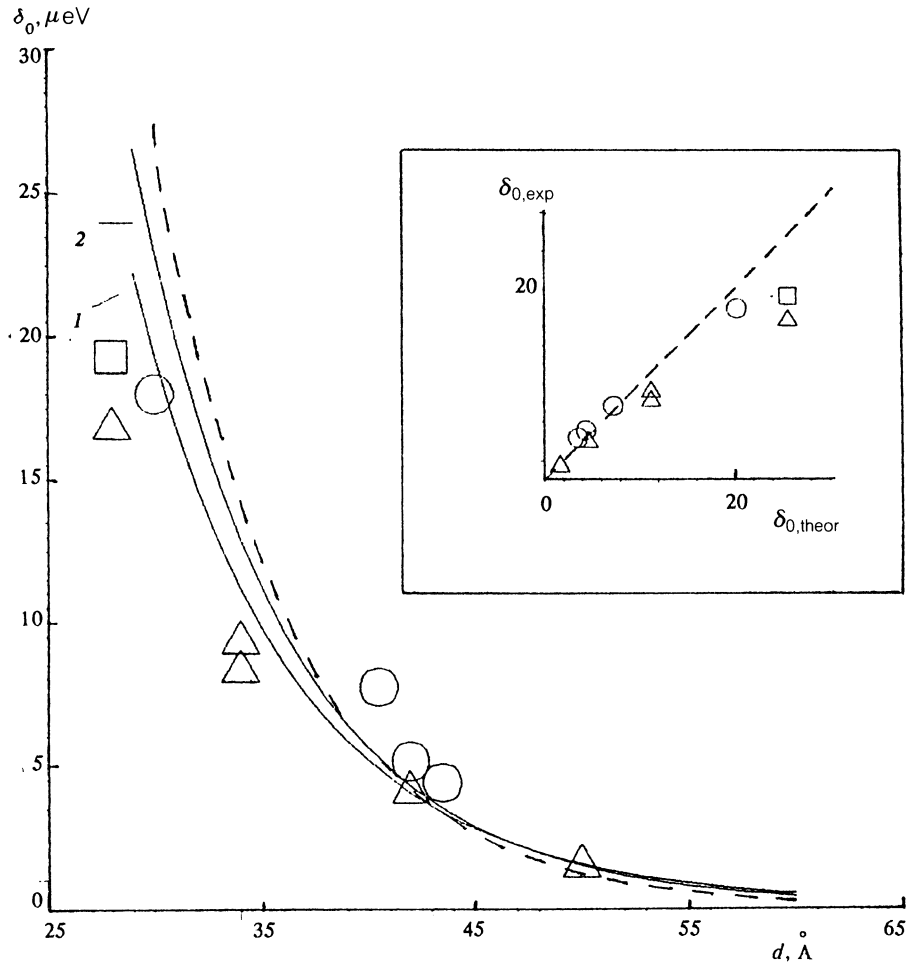

FIG. 1. Axisymmetric exchange splitting δ_0 of the exciton level $e1X$–$hh1(1s)$ as a function of the SL period $d=a+b$. Experiment: △—Ref. 5; □—Ref. 14; and ○—Ref. 15. Theory: solid curves represent the results of calculations in which in (27) a_{2D} was substituted for $a_\perp$ at $a=b$ (curve *1*) and at $a=b/2$ (curve *2*), and the dashed curve represents the results of calculations for a SL with $a=b/2$ at values of $a_\perp$ found by the variational method with the trial functions specified in Eqs. (6) and (7). In the inset the experimental values $\delta_{0,exp}$ measured in a SL with different layer thicknesses a and b are compared with the theoretical values $\delta_{0,theor}$ calculated at $a_\perp = a_{2D}$ for the same thicknesses a and b.

where L is the localization range. Since the exchange-interaction operator (24) is independent of the position of the exciton's center of mass $\mathbf{R}_\perp$, the exchange-interaction constants, including $\delta_2^{(II)}$, are independent of the specific shape of the envelope $f(\mathbf{R}_\perp)$, in view of which the exciton localization parameters are not incorporated in Eqs. (27) and (28). In this connection we note that in a type II superlattice in which two neighboring GaAs and AlAs layers are typical, say, thicker than the other layers by a monolayer, the fine structure of a free quasi-two-dimensional exciton in which the electron is in the typical AlAs layer and the hole in the typical GaAs layer is also described by Eqs. (27) and (28).

If we allow for l–h mixing, the splitting $\delta_1^{(II)}$ of optically inactive states is determined by the expression

$$\delta_1^{(II)} = -\frac{\varepsilon_1}{\varepsilon_0+9\varepsilon_1/4}\delta_0^{(II)} + \left(\varepsilon_0+\frac{5}{2}\varepsilon_1\right)\frac{16a_0^3}{3\pi a_\perp^2} \times \int S^2(z)\phi_e^2\left(z\pm\frac{d}{2}\right)dz. \quad (29)$$

We see that for $t_{l-h}\neq 0$ this splitting can be finite even if the cubic invariant in (24) is ignored.

Estimates show that in calculating $\delta_n^{(II)}$ $(n=0,1,2)$ in type II superlattices the mixing of Γ states to the function (6) can be discarded. In the vicinity of a type I–type II transition this mixing becomes noticeable, and Eqs. (27)–(29) acquire a factor that is the square of the envelope function for Γ states. Note that in contrast to exchange interaction, optical excitation or emission of an $e1X$-$hh1$ exciton occurs only owing to Γ–X mixing.

6. RESULTS OF CALCULATIONS AND COMPARISON WITH EXPERIMENTAL DATA

In calculating the exchange constants $\delta_n^{(II)}$, we employed the following values of the band parameters:

$$m_{hh}=0.45m_0,\quad m_{lh}=0.09m_0,\quad m_{hh1}^\perp=0.15m_0,$$

$$m_x^\parallel=1.8m_0,\quad m_x^\perp=0.2m_0,\quad V_h=0.53\text{ eV},$$

$$\Delta=0.35\text{ eV},\quad \bar{\Delta}=0.46\text{ eV},\quad R=0.9\text{ eV}\cdot\text{Å}^{-1},\quad \kappa=13.$$

The constant ε_1 was set in Eqs. (27) and (28) at zero and the constants t_{l-h} and ε_0 were found from the requirement of best fit to the experimental data on $\delta_0^{(II)}$ and $\delta_2^{(II)}$. It is convenient to determine the value of t_{l-h} from the ratio $\delta_2^{(II)}\delta_0^{(II)}$, which is independent of ε_0 and of $a_\perp$. For the quasi-two-dimensional radius+ $a_\perp$ in (27) and (28) we took either the value $a_\perp = a_{2D}$ (the two-dimensional approximation) or the value determined from the require-

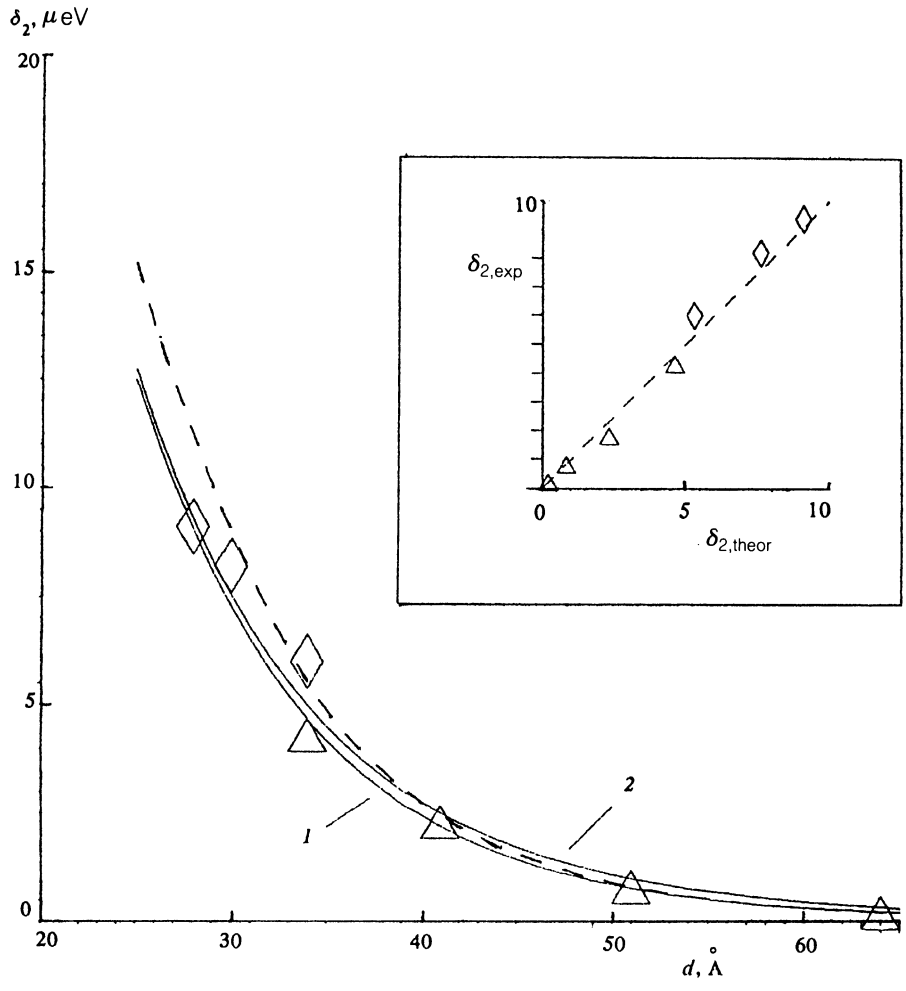

FIG. 2. Anisotropic exchange splitting δ_2 of the emission doublet as a function of the SL period. Experiment: △—Ref. 5; and ◇—Ref. 16. Theory: solid curves represent the results of calculations in which in (28) a_{2D} was substituted for $a_\perp$ with $a=2b/5$ (curve *1*) and with $a=3b/5$ (curve *2*), and the dashed curve represents the results of calculations for a SL with $a=3b/5$ at $a_\perp$ values found by the variational method. In the inset the experimental values $\delta_{2,exp}$ are compared with the theoretical values $\delta_{2,theor}$.

ment that the energy of the exciton with the wave function (9) be minimal (the quasi-two-dimensional approximation), that is, from the minimum condition imposed on the functional

$$\varepsilon_{exc}=\frac{\hbar^2}{2\mu_\perp a_\perp^2}-\int\int\int\frac{e^2}{\kappa[(z_e-z_h)^2+\rho^2]^{1/2}}\frac{2}{\pi a_\perp^2}$$
$$\times\exp\left(-\frac{2\rho}{a_\perp}\right)C^2(z_e)\phi_e^2\left(z_e\pm\frac{d}{2}\right)2\pi\rho\,d\rho\,dz_e\,dz_h. \quad (30)$$

The best fit to the experimental data is achieved with $t_{l\text{-}h}=1.4$ and $\varepsilon_0=54$ meV (the two-dimensional approximation) and $\varepsilon_0=215$ meV (the quasi-two-dimensional approximation). Currently we know of no independent data on the constant ε_0 of exchange interaction between an X-electron and a Γ-hole in GaAs or AlAs, since experiments[5,14-16] have determined the constants $\delta_0^{(II)}$ and $\delta_2^{(II)}$, and the formulas for these quantities contain, in addition to ε_0 and ε_1, band parameters and the Bohr radius of a quasi-two-dimensional exciton.

Figures 1 and 2 depict the theoretical curves representing the dependence of the axisymmetric-exchange-interaction constant δ_0 and the anisotropic-exchange-interaction constant δ_2 on the period d for fixed a/b values. The figures also depict the known experimental values of δ_0 and δ_2. We see that, in accordance with the experimental data, both δ_0 and δ_2 sharply increase as the period gets smaller and depend weakly on a/b for a fixed value of $a+b$. Since in the $e1X$–$hh1$ exciton the electron and hole are localized chiefly in different layers, allowing for the spatial distribution along the z axis of the electron and hole density, we find that the Coulomb energy, defined by the second term on the right-hand side of (30), differs considerably from the Coulomb energy $-e^2/\kappa a_{2D}$ for a two-dimensional exciton. This explains the difference in values of parameter ε_0 found in the two-dimensional and quasi-two-dimensional approximations. Comparison of the solid curves and the dashed curve in Fig. 1 shows that the choice of approximation affects the dependence of the exchange splitting on layer thickness only moderately.

Note that in a more accurate calculation of the wave function of a localized $e1X$–$hh1$ exciton we must allow for the effect of Coulomb attraction on the motion of the electron and hole along the z axis. As a result the shape of the single-particle functions $\psi_{hh1}(z_h)$ and $u_0(z_e)$ and $v_0(z_e)$ changes and their maxima shift toward the heterojunction. Hence, we should expect that an exact calculation will lead to values of $\delta_0^{(II)}$ and $\delta_2^{(II)}$ that lie within the limits specified by the two-dimensional and quasi-two-dimensional approximations.

Finally, it is worth noting that a good fit to the experimental data has also been obtained for the constant $\delta_1^{(II)}$ at $\varepsilon_1=0$ but with allowance for l–h mixing. For instance, for superlattices with thickness ratios of 17 Å/17 Å (the G485 sample), 22 Å/19 Å, and 23 Å/28 Å we obtained the experimental values $\delta_1^{(II)}=0.5$, 0.4, and 0.15 μeV, while calculations yielded $\delta_1^{(II)}=0.57$, 0.34, and 0.15 μeV.

Thus, the theory of electron–hole exchange interaction developed here yields a satisfactory explanation of the fine structure of excitonic levels studied in GaAs/AlAs superlattices.

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[1] G. Danan, B. Etienne, F. Mollot *et al.*, Phys. Rev. B **35**, 6207 (1987).
[2] E. Finkman, M. D. Sturge, M.-H. Meynadier *et al.*, J. Lumin. **39**, 57 (1987).
[3] H. W. van Kesteren, E. C. Cosman, P. Dawson *et al.*, Phys. Rev. B **39**, 13 426 (1989).
[4] D. Scalbert, J. Cernogora, Benoit a la Guillaume *et al.*, Solid State Commun. **70**, 945 (1989).
[5] H. W. van Kesteren, E. C. Cosman, W. A. J. A. van der Poel, and C. T. Foxon, Phys. Rev. B **41**, 5283 (1990).
[6] S. Permogorov, A. Naumov, C. Gourdon, and P. Lavallard, Solid State Commun. **74**, 1057 (1990).
[7] E. L. Ivchenko, V. P. Kochereshko, A. Yu. Naumov *et al.*, Superlatt. Microstruct. **10**, 497 (1991).
[8] I. L. Aleĭner and E. L. Ivchenko, Pis'ma Zh. Eksp. Teor. Fiz. **55**, 662 (1992) [JETP Lett. **55**, 692 (1992)].
[9] I. L. Aleĭner and E. L. Ivchenko, Fiz. Tekh. Poluprovodn. **27**, 594 (1993) [Semiconductors **27**, 330 (1993)].
[10] E. L. Ivchenko, V. P. Kochereshko, P. S. Kop'ev *et al.*, Solid State Commun. **70**, 529 (1989).
[11] T. Ando and H. Akera, Phys. Rev. B **40**, 11 609 (1989).
[12] G. L. Bir and G. E. Pikus, *Symmetry and Strain–Induced Effects in Semiconductors*, Wiley, New York (1974).
[13] G. E. Pikus and G. L. Bir, Zh. Eksp. Teor. Fiz. **60**, 195 (1971) [Sov. Phys. JETP **33**, 108 (1971)].
[14] J. M. Trombetta, T. A. Kennedy, D. Gammon *et al.*, in *Proc. 20th Int. Conf. Phys. Semicond.*, Vol. 2, Salonika (1990), p. 1361. Wiley, New York (1974).
[15] P. G. Baranov, P. Lavallard, R. Planel, and N. G. Romanov, Superlatt. Microstruct. (to be published) (1993).
[16] C. Gourdon and P. Lavallard, Phys. Rev. B **46**, 4644 (1992).

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