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Theory of exciton energy levels in multiply periodic systems

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Abstract. A general theoretical formalism is developed for evaluating the lowest energy levels of an exciton in a system which is of finite size, but multiply periodic, in one dimension, whilst being of infinite extent in the other two directions. A particular application of the formalism to a Kronig–Penney type I multiquantum well-superlattice structure is made and the significance of the results described.

1. Introduction

The aim of the present paper is to describe a mathematical technique which effects great simplifications in the calculation of the lowest energy bound states of an interacting 'two-body' system (namely, an electron and hole), for a solid-state system which possesses translational symmetry in the x–y plane (with a period $\sim$ few Å), together with a periodically varying potential in the z-direction (with a period D typically $\sim$100 Å). Such systems have been actively studied in recent years under the general title of low-dimensional structures [1–3]. From the experimental viewpoint, the optical properties of these systems are determined largely by the lowest energy bound states of the electron and the hole (i.e. the so-called 'exciton' energy levels). If the potential energy variation in the z-direction is restricted to a single period, we have what is usually referred to as a single quantum well, for which an extensive number of calculations of the exciton energy levels have been made in terms of the envelope function approximation [4–7]. On the other hand, a system with a large number of periods in the z-direction (say N) is referred to as a multiquantum well. There appears to be a dearth of calculations of the energy levels of excitons in such systems. One reason for this is that the N wells of a multiquantum well system can, for wide choices of the period D, act like N independent single wells. However there are other ranges of D where the energy levels in adjacent well interact appreciably with one another, giving rise to what is usually called a 'superlattice'. The present paper employs the envelope function approximation, and within this approach develops a general technique based on the variational principle to evaluate the lowest energy levels of excitons for both multiquantum wells and for superlattices. The general formalism applicable to any periodic structure is first described, before application is made to a particular type of structure called a type I superlattice.

2. Theoretical model

The model assumes the validity of the envelope function approximation, which takes the wavefunction of the interacting system of an electron and hole in a semiconductor

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to be written as the product of a Bloch function appropriate to the bottom of the associated (electron) conduction or (hole) valence band states together with an envelope function describing the relative electron–hole motion. It can be shown that the latter satisfies the Schrödinger time-independent wave equation appropriate to a Hamiltonian having the form [7]

$$H = H_1(\mathrm{e}) + H_2(\mathrm{h}) + H_3(\perp) + H_4 \tag{2.1}$$

where $H_1(e)$ describes the periodic one-electron conduction band Hamiltonian appropriate to the z-direction

$H_2(h)$ is the equivalent Hamiltonian for the hole

$H_3(\perp)$ describes the kinetic energy of the relative motion of the electron and hole in the x-y plane

H_4 is the Coulombic potential energy interaction term of the electron and hole.

The present paper is *not* concerned with the limits of validity of the envelope-function approximation rather it addresses the problem of solving the resulting Schrödinger equation

$$H\Phi = E\Phi \tag{2.2}$$

for the envelope wavefunctions Φ and the associated energy levels E. It will be noted that the Hamiltonian in (2.1) does not contain a kinetic energy term for the motion of the centre-of-mass of the electron and hole. This is because we are interested solely in the lowest energy exciton states, which correspond to a wavevector $\boldsymbol{K}$ of zero for the centre-of-mass motion.

The envelope function is taken to have the form

$$\Phi = \varphi_\mathrm{e}(z_\mathrm{e})\varphi_\mathrm{h}(z_\mathrm{h})\varphi \tag{2.3}$$

where $\varphi_\mathrm{e}(z_\mathrm{e})[\varphi_\mathrm{h}(z_\mathrm{h})]$ is chosen to be an eigenfunction of the periodic one-electron Hamiltonian $H_1(\mathrm{e})[H_2(\mathrm{h})]$, i.e.

$$H_1(\mathrm{e})\varphi_\mathrm{e}(z_\mathrm{e}) = E_\mathrm{e}\varphi_\mathrm{e}(z_\mathrm{e}) \tag{2.4}$$

$$H_2(\mathrm{h})\varphi_\mathrm{h}(z_\mathrm{h}) = E_\mathrm{h}\varphi_\mathrm{h}(z_\mathrm{h}) \tag{2.5}$$

and φ is a (variational) wavefunction whose form will be chosen later. Given the specific form of $H_1(\mathrm{e})$ and $H_2(\mathrm{h})$ the solution of equations (2.4) and (2.5) is a straightforward task. For the general development below we will assume that E_e, E_h, $\varphi_\mathrm{e}(z_\mathrm{e})$ and $\varphi_\mathrm{h}(z_\mathrm{h})$ are known with both $\varphi_\mathrm{e}(z_\mathrm{e})$ and $\varphi_\mathrm{h}(z_\mathrm{h})$ being normalized over the multiquantum-well region. Substitution of (2.3) into (2.2) gives

$$E = \frac{\int \Phi^* H \Phi \, \mathrm{d}\tau}{\int \Phi^* \Phi \, \mathrm{d}\tau}. \tag{2.6}$$

In order to produce further simplifications in the expression for E, we note that the function φ describes the relative motion of the electron and hole, and as such can be taken to be a function of the three variables $x_\perp = |x_\mathrm{e} - x_\mathrm{h}|$, $y_\perp = |y_\mathrm{e} - y_\mathrm{h}|$ and $a = |z_\mathrm{e} - z_\mathrm{h}|$. Thus the denominator has the form

$$\int |\varphi_\mathrm{e}(z_\mathrm{e})|^2 \, \mathrm{d}z_\mathrm{e} |\varphi_\mathrm{h}(z_\mathrm{h})|^2 \, \mathrm{d}z_\mathrm{h} |\varphi|^2 \, \mathrm{d}x_\perp \, \mathrm{d}y_\perp. \tag{2.7}$$

Since the x-y plane is taken to be of infinite extent the integral

$$\int |\varphi|^2 \, \mathrm{d}x_\perp \, \mathrm{d}y_\perp$$

will be a function of $a = |z_\mathrm{e} - z_\mathrm{h}|$ only, whatever the precise form of φ, i.e.

$$F(a) = \int |\varphi|^2 \, \mathrm{d}x_\perp \, \mathrm{d}y_\perp .$$

Hence (2.7) can be recast in the form

$$\int |\varphi_\mathrm{e}(z_\mathrm{e})|^2 \, \mathrm{d}z_\mathrm{e} |\varphi_\mathrm{h}(z_\mathrm{h})|^2 \, \mathrm{d}z_\mathrm{h} \, F(a). \tag{2.8}$$

Bearing in mind the probability interpretation of the wavefunction we can rewrite (2.8) as

$$\int p(a) F(a) \, \mathrm{d}a \tag{2.9}$$

where $p(a)$ is the (uncorrelated) probability of finding an electron and a hole separated by distance a.

Analogous simplifications can be effected in the numerator in (2.6). To this end we note the kinetic energy operators in $H_1(\mathrm{e})$ and $H_2(\mathrm{h})$ will act on the function φ. Since, whatever the precise form of φ, the following relation is valid

$$\frac{\partial \varphi}{\partial z_\mathrm{e}} = \frac{-\partial \varphi}{\partial z_\mathrm{h}}$$

the extra term in the numerator, in addition to the one-electron energies, E_e and E_h that arise from the kinetic energy operators along the z-direction, can be readily shown to have the form

$$\frac{\hbar^2}{2\mu} \int |\varphi_\mathrm{e}(z_\mathrm{e})|^2 \, \mathrm{d}z_\mathrm{e} |\varphi_\mathrm{h}(z_\mathrm{h})|^2 \, \mathrm{d}z_\mathrm{h} \left| \frac{\partial \varphi}{\partial z_\mathrm{e}} \right|^2 \mathrm{d}x_\perp \, \mathrm{d}y_\perp \tag{2.10}$$

with

$$\frac{1}{\mu} = \frac{1}{(m_\mathrm{e})_z} + \frac{1}{(m_\mathrm{h})_z}$$

where $(m_e)_z[(m_\mathrm{h})_z]$ is the electron [hole] effective mass in the z-direction.

Again, in an obvious notation

$$G(a) = \int \left| \frac{\partial \varphi}{\partial z_\mathrm{e}} \right|^2 \mathrm{d}x_\perp \, \mathrm{d}y_\perp \tag{2.11}$$

and (2.10) has the form

$$\frac{\hbar^2}{2\mu} \int p(a) G(a) \, \mathrm{d}a. \tag{2.12}$$

Utilizing all the previous equations enables us to recast (2.6) in the form

$$E = E_\mathrm{e} + E_\mathrm{h} + \frac{\hbar^2}{2\mu} \frac{\int p(a) G(a) \, \mathrm{d}a}{\int p(a) F(a) \, \mathrm{d}a} + \frac{\int \Phi(H_3(\perp) + H_4) \Phi \, \mathrm{d}z}{\int p(a) F(a) \, \mathrm{d}a}. \tag{2.13}$$

Further simplification of (2.13) can only be effected by employing a specific choice for the function φ. In order to illustrate, we will consider the 1*s*-state of an exciton. For this situation the function φ is taken to have the form

$$\varphi = \exp\left(\frac{-r'}{\lambda}\right) \tag{2.14}$$

where $(r')^2 = (x_e - x_h)^2 + (y_e - y_h)^2 + (1-\beta^2)(z_e - z_h)^2$

i.e. $$(r')^2 = r_\perp^2 + (1-\beta^2)a^2 \tag{2.15}$$

where β and λ are variational parameters chosen so as to maximize the binding energy of the exciton. The key feature of the present analysis which we wish to demonstrate is concerned with effecting great simplifications in the integrations over the x-y plane. To this end consider the Hamiltonian

$$H_3(\perp) = \frac{-\hbar^2}{2m_\perp}\left(\frac{\partial^2}{\partial x_\perp^2} + \frac{\partial^2}{\partial y_\perp^2}\right)$$

where $m_\perp$ is the reduced mass in the x-y plane. In the evaluation of the numerator in (2.13) we will meet integrals of the following type

$$\int_0^\infty \frac{r_\perp^2}{(r')^2} \exp\left(\frac{-2r'}{\lambda}\right) r_\perp \, \mathrm{d}r_\perp . \tag{2.16}$$

To simplify this integral we first make the scale change (see (2.15))

$$r_\perp = (1-\beta^2)^{1/2} a r'_\perp$$

followed by $r'_\perp = \sinh\theta$ to give

$$\int_0^\infty (1-\beta^2)a^2 \sinh^2\theta \tanh\theta \exp[(-2/\lambda)(1-\beta^2)^{1/2} a \cosh\theta] \, \mathrm{d}\theta. \tag{2.17}$$

In turn we then change variable to $x = \mathrm{e}^{-\theta}$ thus yielding

$$\frac{(1-\beta^2)a^2}{4} \int_0^1 \frac{1}{x}\left(\frac{1}{x} - x\right)^3 \left(\frac{1}{x} + x\right)^{-1} \exp[(-(1-\beta^2)^{1/2}a)/\lambda(1/x + x)] \, \mathrm{d}x. \tag{2.18}$$

There are two points to note about the result in (2.18). First it is a function of $a = |z_e - z_h|$—thus making it of the same functional form as the $G(a)$ appearing in (2.11). This means that when the integration over z_e and z_h is subsequently carried out, the resulting integral can again be cast into the same general form as that in (2.12). Secondly, the range of integration in (2.18) extends from 0 to 1 only—thus enabling an easy and accurate numerical evaluation of it to be made on a computer for a given choice of β, a and λ. This same technique permits all the integrals to be cast into the same general form without the need for reverting to a series expansion as is normally done for example in evaluating H_4 [4–6]. As a final example to illustrate this feature we consider the term in the numerator on the right-hand side of (2.13) associated with H_4. The resulting integral involves the following term

$$\int_0^\infty \frac{1}{r} \mathrm{e}^{-2r'/\lambda} r_\perp \, \mathrm{d}r_\perp \tag{2.19}$$

where

$$r^2 = r_\perp^2 + a^2. \tag{2.20}$$

From (2.20) we have, for a fixed value of a,

$$r\,\mathrm{d}r = r_\perp\,\mathrm{d}r_\perp$$

and (2.19) becomes

$$\int_a^\infty \mathrm{e}^{-2r'/\lambda}\,\mathrm{d}r. \tag{2.21}$$

From (2.15) we have

$$r'^2 = r^2 - \beta^2 a^2.$$

We therefore make the scale change

$$r = \beta a r_1$$

followed by $r_1 = \cosh\theta$ to given in place of (2.21)

$$\int_{\cosh^{-1}(1/\beta)}^\infty \beta a \sinh\theta \exp[(-2\beta a)/\lambda \sinh\theta]\,\mathrm{d}\theta.$$

Introducing $x = \mathrm{e}^{-\theta}$ gives finally

$$\frac{\beta a}{2}\int_0^{(1/\beta)-\sqrt{(1/\beta^2)-1}} \frac{1}{x}\left(\frac{1}{x}-x\right)\exp[(-\beta a)/\lambda(1/x-x)]\,\mathrm{d}x.$$

Note again that the range of integration satisfies $0 \leqslant x \leqslant 1$, since (see (2.15)) β satisfies $0 \leqslant \beta \leqslant 1$, with $\beta = 0$ being a 3D exciton and $\beta = 1$ representing a 2D exciton.

In summary we see that the present approach reduces the problem of calculating exciton binding energies to an evaluation of a series of integrals of the type

$$I = \int p(a)G(a)\,\mathrm{d}a$$

where $G(a)$ involves numerically integrating a given function over the variable x with x satisfying $0 \leqslant x \leqslant 1$. The entity $p(a)$ represents the (uncorrelated) probability of finding an electron and hole separated by a distance a. To evaluate the $p(a)$s, we simply solve the one-electron Schrödinger equations (2.4) and (2.5), normalize the resulting wavefunctions φ_e and φ_h and then (numerically if necessary) evaluate integrals of the type

$$\int |\varphi_e(z_e)|^2 |\varphi_h(z_e + a)|^2\,\mathrm{d}z_e. \tag{2.22}$$

It is to be stressed that within the present formalism the only difference betwen the various possible periodic structures whether they be sawtooth, rectangular, parabolic, etc., is in the form of the $p(a)$s. Similarly, for a given structure, the only difference between a multiquantum well and a superlattice is the variation in the value of the $p(a)$s with the period D.

3. Application to a type I superlattice

Figure 1 depicts the potential well structure for what is termed a type I superlattice. The number of periods can be extended indefinitely. In order to illustrate the present

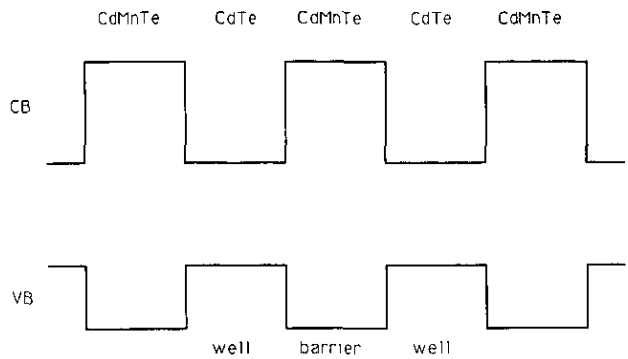

Figure 1. Conduction band (CB) and valence band (VB) alignment at $\Gamma = 0$ for a typical CdTe-CdMnTe type I superlattice.

technique we will evaluate the $1s$ and $2s$ exciton energy levels for a simple Kronig-Penney type superlattice. (Note, the inclusion of more complicated band structure effects manifests itself solely in the evaluation of the $p(a)$s, and involves replacing the one-electron wavefunctions $\varphi_e(z_e)$ and $\varphi_h(z_h)$ employed below by the corresponding values given by solving, for III-V or II-VI compounds for example, the standard one-electron 8×8 matrix given in the literature [3].) The boundary conditions for the latter are well known and involve matching wavefunctions and their derivatives at various points [8]. Since, in the authors' laboratory, we are interested in II-VI compounds we have employed, for the purpose of illustration, effective masses and dielectric constants appropriate to the CdTe ($Cd_{1-x}Mn_xTe$) system.

For the particular case of a Kronig-Penney type superlattice, the expression for the $p(a)$s can be obtained in a closed analytical form. However they are not reproduced here since they are extremely lengthy. Furthermore, they offer no advantage from the computational viewpoint, from the values obtained by straightforward numerical integration of (2.22). For the $1s$ wavefunctions the trial function in (2.14) was employed whilst for the $2s$ state the following function was used:

$$\varphi_{2s} \sim \left(1 - \frac{\alpha r'}{\lambda_2}\right) \exp\left(\frac{-r'}{\lambda_2}\right). \tag{3.1}$$

The parameters α and λ_2 for the $2s$-state are not wholly independent of the λ parameter determined for the $1s$ state, since the total wavefunction Φ (see (2.3)) for these two states must be orthogonal to one another.

To be definite we choose for the $CeTe/Cd_{1-x}Mn_xTe$ system a value of $x = 0.1$. The resulting binding energies for the $1s$ and $2s$ excitons are shown in figure 2. It is evident from this figure that with increasing well width the exciton binding energy rises to a maximum before decreasing again at larger well widths. Both the $1s$ and $2s$ binding energies peak at approximately the same value of well width ~ 100 Å, and the binding energy of the $1s$ state is approximately four to five times that of the $2s$ state. The relative value of the binding energy does not vary much with well width for either the $1s$ or $2s$ states. Furthermore, the magnitudes of these binding energies, at any particular value of the well width, is relatively insensitive to the fraction of the total energy band offset (i.e. the difference in the band gaps between CdTe and $Cd_{1-x}Mn_xTe$) that is assigned to the valence band.

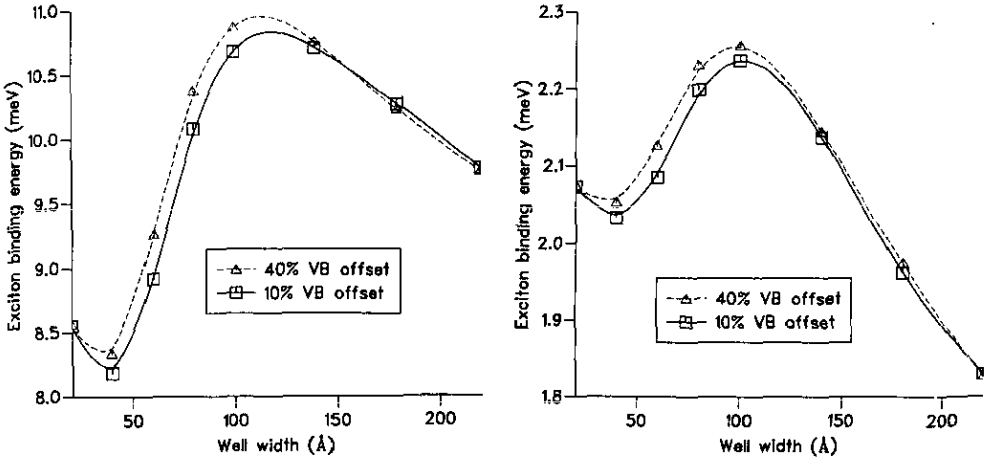

Figure 2. Binding energy as a function of well width for a CdTe-CdMnTe superlattice with 10% and 40% of the offset in the valence band for (*a*) 1*s*, (*b*) 2*s* exciton.

We consider next a related question, namely the value of the radiative transition probabilities (or, a closely related entity, the oscillator strength f). It is well known that, in terms of the previous notation in (2.3)

$$f \propto \left| \int \Phi(\boldsymbol{r}, \boldsymbol{r})\, \mathrm{d}^3\boldsymbol{r} \right|^2 .$$

The resulting values for the oscillator strength of the 1*s* and 2*s* excitons calculated in this way are shown in figure 3 (which employ the same arbitrary units). The behaviour of the oscillator strengths is similar to that of the binding energies in that they increase with the well width, peak in the same region ~100 Å and then decrease again. However they differ from the binding energy curves in comparative terms in three important ways. First the oscillator strengths of the 2*s* exciton are typically two orders of magnitude less than those of the 1*s* exciton. The second difference is that the relative change in

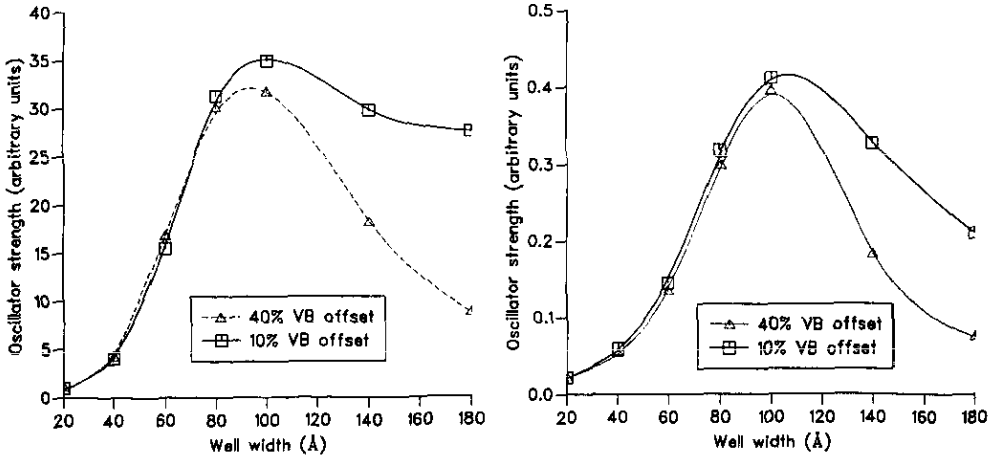

Figure 3. Relative oscillator strength as a function of well width for a CdTe-CdMnTe superlattice with 10% and 40% of the offset in the valence band for (*a*) 1*s*, (*b*) 2*s* exciton.

the oscillator strengths with varying well width is much greater than the corresponding changes in the exciton energies. Thirdly, for the wider wells, there is a difference involving a factor of two to three between the oscillator strengths for the different values of the offset. These results are a good illustration of the fact that the Schrödinger equation, being equivalent to the solution of a variational problem, has energy eigenvalues that are insensitivite to slight changes in the wavefunction. However other physical properties, such as the oscillator strength, are far more sensitive measures of such variations.

4. Conclusion

We have described a method for calculating the energy levels of an exciton whose wavefunction can be written in a product form (see (2.3)). The formalism is perfectly general and can be applied to a 1D potential of arbitrary shape. For the purpose of illustration we have chosen a type I superlattice and evaluated the well width dependence of both the exciton binding energy and the oscillator strength for a $1s$ and a $2s$ exciton. The general form of the curves are in agreement with those described in the literature [9, 10] for related physical systems.

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