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LETTER TO THE EDITOR

Excitons in superlattices

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Abstract. A simple, non-variational approach for calculating exciton binding energies in superlattices is developed. The approach is sufficiently versatile that it applies in the limits of strongly coupled and isolated quantum wells, *and* in the presence of external fields. Excellent agreement is obtained with experimental binding energies in GaAs/GaAlAs using only bulk $\boldsymbol{k}\cdot\boldsymbol{p}$ parameters as input.

Many experimental and theoretical studies have been reported of excitons in single quantum wells [1]. Increasing attention is now being paid to the properties of excitons in superlattices with finite barrier thicknesses [2–5], such that the exciton can spread over several superlattice periods. In particular, experimental values of superlattice exciton binding energies both with [2] and without [3] external electric fields have recently been reported. Theoretical studies of excitons in superlattices have however primarily utilised variational 'particle-in-a-box' approaches or the computationally intensive $\boldsymbol{k}$-space sampling technique of Chu and Chang [6].

In this letter we present a simple and versatile non-variational approach for calculating superlattice binding energies based on the crystal coordinate representation (CCR) using a basis consisting of *superlattice* Wannier functions. The formalism underlying the present approach exploits the 3D superlattice periodicity, as in the case of the one-electron f-sum rule [7], and is applicable to semiconductor superlattices (types I, II and III) with arbitrary layer widths. The exciton equation in the CCR representation yields a 1D tight-binding equation involving the growth axis. An approximate solution yields excellent agreement with recently measured binding energies in GaAs/GaAlAs superlattices as a function of (i) layer widths spanning the range from strongly coupled to isolated quantum wells [3] and (ii) electric field strength [2]. Preliminary results from this work have been briefly reported [8]. The effects of other (e.g. magnetic) fields will be considered in a future publication [9].

The eigenstates of the superlattice single-particle Hamiltonian H_{SL} are defined for band L at wavevector $\boldsymbol{K}$ by

$$H_{SL}\,|L\boldsymbol{K}\rangle = E_L(\boldsymbol{K})\,|L\boldsymbol{K}\rangle. \tag{1}$$

As in the bulk, the superlattice Wannier function at *superlattice* lattice vector $\boldsymbol{R} = (X, Y, Z)$ is defined as

$$\langle \boldsymbol{r}|L\boldsymbol{R}\rangle = (\Omega/8\pi^3)^{1/2}\int \mathrm{d}\boldsymbol{K}\,\mathrm{e}^{-\mathrm{i}\boldsymbol{K}\cdot\boldsymbol{R}}\langle \boldsymbol{r}|L\boldsymbol{K}\rangle \tag{2}$$

where Ω is the normalisation volume. The superlattice growth ($\perp$) axis is taken to be

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the Z axis while the in-plane ($\|$) directions are described by X and Y. The superlattice period in the $\perp$ ($\|$) direction is d (a), where d is typically of the order of 100 Å while $a \simeq$ 6 Å. The superlattice Wannier function will have a spatial extent of the order of d (a) in the $\perp$ ($\|$) direction [9].

The many-body Hamiltonian is given by

$$H = \sum_i H_{SL}(\boldsymbol{r}_i) + \sum_{j>i} v(\boldsymbol{r}_i - \boldsymbol{r}_j) \tag{3}$$

where the latter term is the Coulomb interaction. We are only interested in electron–hole excitations and expand the exciton wavefunction in a basis of two-particle Wannier states $|L\boldsymbol{R} + \boldsymbol{R}'; L'\boldsymbol{R}'\rangle$ each consisting of a symmetrised product of electron (hole) Wannier functions [10] for band L (L') centred at $\boldsymbol{R} + \boldsymbol{R}'$ ($\boldsymbol{R}'$). To simplify notation we only consider two superlattice bands although the formalism can be easily generalised. The total exciton state $|\Psi\rangle$ at zero exciton wavevector (i.e. the optically active exciton) can be written [10] as

$$|\Psi\rangle = (\Omega/8\pi^3)^{1/2} \sum_{\boldsymbol{R}} U(\boldsymbol{R}) \sum_{\boldsymbol{R}'} |L\boldsymbol{R}' + \boldsymbol{R}; L'\boldsymbol{R}'\rangle \tag{4}$$

where $U(\boldsymbol{R})$ is the exciton CCR wavefunction. The sum over $\boldsymbol{R}'$ includes all two-particle states with a given electron–hole separation $\boldsymbol{R}$ while the sum over $\boldsymbol{R}$ accounts for all possible electron–hole separations. Evaluating the many-body Hamiltonian of (3) in the two-particle basis leads to a set of difference equations [10]

$$\sum_{\boldsymbol{R}'}\left((\Omega/8\pi^3)\int \mathrm{d}\boldsymbol{K}\, \mathrm{e}^{\mathrm{i}\boldsymbol{K}\cdot(\boldsymbol{R}-\boldsymbol{R}')}(E_L(\boldsymbol{K}) - E_{L'}(\boldsymbol{K}))\right)U(\boldsymbol{R}') - V(\boldsymbol{R})U(\boldsymbol{R}) = EU(\boldsymbol{R}). \tag{5}$$

Here E is the excitation energy of the superlattice and $V(\boldsymbol{R})$ is the direct Coulomb term [11] given by

$$V(\boldsymbol{R}) = \int\int |\langle \boldsymbol{r}_\mathrm{e}|L\boldsymbol{R}\rangle|^2 \frac{e^2}{\varepsilon|\boldsymbol{r}_\mathrm{e} - \boldsymbol{r}_\mathrm{h}|} |\langle \boldsymbol{r}_\mathrm{h}|L'0\rangle|^2 \,\mathrm{d}\boldsymbol{r}_\mathrm{e}\,\mathrm{d}\boldsymbol{r}_\mathrm{h} \tag{6}$$

where $\boldsymbol{r}_\mathrm{e}$ ($\boldsymbol{r}_\mathrm{h}$) defines the electron (hole) coordinates and ε is the static dielectric constant.

To evaluate (5) for the superlattices of interest, we write

$$E_L(\boldsymbol{K}) - E_{L'}(\boldsymbol{K}) = \frac{\hbar^2 K_\|^2}{2\mu_\|} + E_\mathrm{g} - \sum_{n=1}^{\infty} 2W(n)(\cos(nK_\perp d) - 1) \tag{7}$$

where $\mu_\|$ is the parallel reduced mass and E_g is the $\boldsymbol{K} = \boldsymbol{0}$ gap between bands L and L'. The parabolic approximation has been made in the parallel direction. The expansion of the perpendicular dispersion as a cosine series with coefficients $W(n)$ exploits the approximate inversion symmetry of superlattice energy bands under $K_\perp \rightarrow -K_\perp$. The equivalent of the effective mass approximation is then made in the parallel direction where the exciton radius (typically 100 Å in the bulk semiconductors of interest) is much greater than the underlying superlattice unit cell size ($a \simeq 6$ Å). The lattice vector coordinates X, Y can therefore be treated as continuous variables. Notice that the effective mass approximation *cannot* be made in the growth direction since d is of the

order of the exciton radius. Using (7) and the above approximations, equation (5) yields a set of coupled differential equations

$$\left[\frac{-\hbar^2}{2\mu_\parallel}\left(\frac{\partial^2}{\partial X^2}+\frac{\partial^2}{\partial Y^2}\right)-V(X,Y,jd)\right]U(X,Y,jd)-\sum_{n=1}^{\infty}W(n)\big(U(X,Y,(j+n)d)$$

$$+U(X,Y,(j-n)d)\big)=\left(\varepsilon-\sum_{n=1}^{\infty}2W(n)\right)U(X,Y,jd) \tag{8}$$

where $\varepsilon=(E-E_g)$ represents the exciton binding energy. The two possible cases of (8) are as follows.

(i) $W(n)=0$ in (8) for all n, which is characteristic of a thick barrier limit in which the electron and hole cannot tunnel between adjacent wells. Equation (8) becomes

$$\left[\frac{-\hbar^2}{2\mu_\parallel}\left(\frac{\partial^2}{\partial X^2}+\frac{\partial^2}{\partial Y^2}\right)-V(X,Y,jd)\right]U_0^m(X,Y,jd)=\varepsilon_j^{0,m}U_0^m(X,Y,jd) \tag{9}$$

which yields the mth (excited state) exciton solution for a given electron–hole separation of j periods. The corresponding binding energies $\varepsilon_j^{0,m}$ for a given j are analogous to the energy levels of a hydrogenic atom. For convenience we can represent the eigenstate of (9) for a given j and m in terms of an orthonormal basis $\{\hat{e}_{X,Y,jd}\}$ corresponding to the superlattice lattice points; hence

$$\Psi_{ex}^{0,j,m}=\sum_{X,Y,j'}U_0^m(X,Y,j'd)\delta_{jj'}\hat{e}_{X,Y,j'd}.$$

(ii) $W(n)\neq 0$; here the exciton is characterised by a distribution of js. The eigenstate of (8) can therefore be represented as

$$\Psi_{ex}=\sum_{j,m}c_j^m\Psi_{ex}^{0,j,m}. \tag{10}$$

It then follows formally from (8) that [9]

$$\sum_{j,m}\left[\left(\varepsilon_j^{0,m}-\varepsilon+\sum_{n=1}^{\infty}2W(n)\right)\delta_{jj'}\delta_{mm'}-\sum_{n=1}^{\infty}\tilde{W}_{j'j}^{m'm}(n)\delta_{j'j\pm n}\right]c_j^m=0 \tag{11}$$

where

$$\tilde{W}_{j'j}^{m'm}(n)=W(n)\int\!\!\int \mathrm{d}X\,\mathrm{d}Y\,(U_0^{m'}(X,Y,j'd))^*U_0^m(X,Y,jd).$$

Equation (11) is identical to a 1D tight-binding equation where $\varepsilon_j^{0,m}$ is the on-site energy at site j and $\tilde{W}_{j'j}^{m'm}(n)$ is the hopping term. Equation (11) is the main result of the formalism for the zero-field binding energy ε and is applicable to superlattices of arbitrary width.

We now solve (11) approximately to compare with the experimental results for GaAs/GaAlAs [3]. Only the nearest-neighbour hopping $\tilde{W}_{j'j}^{m'm}(1)$ will be retained since we are mostly interested in superlattices with barrier thicknesses greater than 30 Å. In order to obtain values of $\varepsilon_j^{0,m}$, the potential function $V(X,Y,jd)$ needs to be specified (see (6)). Explicit forms for the electron and hole Wannier functions ($\langle \boldsymbol{r}_e|\boldsymbol{LR}\rangle$ and $\langle \boldsymbol{r}_h|L'\boldsymbol{0}\rangle$ respectively) must therefore be used. Since the electron–hole Coulomb interaction is slowly varying on the atomic scale a, the rapidly varying portion of the superlattice Wannier functions in (6) can be integrated out [9], leaving an envelope function

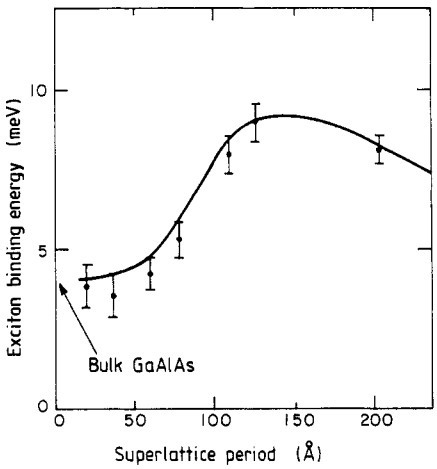

Figure 1. Binding energies of the heavy-hole (C1-HH1) exciton versus superlattice period d for $(d/2)$ GaAs/$(d/2)$ $Ga_{0.7}Al_{0.3}As$. Full curve: present theory; experimental points: from [3].

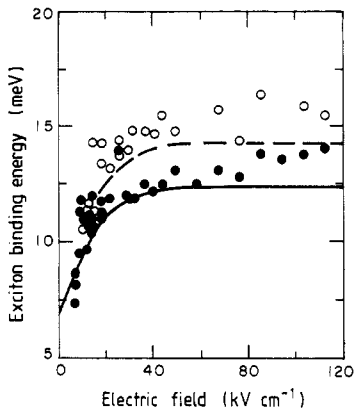

Figure 2. Experimental [2] and theoretical exciton binding energies for the 40 Å GaAs/40 Å $Ga_{0.65}Al_{0.35}As$ superlattice versus electric field strength F. Binding energies of the heavy-hole (C1-HH1) exciton are shown by the full curve (theory) and full circles (experiment). Light-hole (C1-LH1) exciton binding energies are shown by the broken curve (theory) and open circles (experiment).

modulation of each Wannier function on the scale of d (a) in the $\perp$ ($\|$) direction. We then assume for simplicity that the charge distribution associated with the electron (hole) Wannier function is well described by a uniformly charged rod of X–Y cross section equal to $a \times a$ and length l_e (l_h), where l_e (l_h) corresponds to the spatial extent of the electron (hole) Wannier function in the growth direction. We take l_e (l_h) to be equal to the well width plus twice the electron (hole) envelope function decay length in the barrier layers. This simplification leads to an analytic expression for $V(X, Y, jd)$. The resulting nearest-neighbour tight-binding equation is then easily solved. Values of the electron and hole bandwidths and the superlattice in-plane masses [12] are obtained using the envelope function approach of [7] which accounts for valence-band mixing. The bulk $\boldsymbol{k} \cdot \boldsymbol{p}$ input parameters for GaAlAs are the same as in [7].

Figure 1 compares the experimental [3] and present theoretical binding energies for 1s excitons formed from the lowest conduction band C1 and the topmost valence band HH1 in the $(d/2)$ GaAs/$(d/2)$ $Ga_{0.7}Al_{0.3}As$ superlattice at low temperature. The excellent agreement between theory and experiment implies that the simplifying assumptions made in the previous paragraph are reasonable. For small d, the superlattice exciton binding energy is nearly equal to that of the bulk alloy $Ga_{0.85}Al_{0.15}As$ chosen to have the same Al concentration as the superlattice. The exciton CCR wavefunction $U(X, Y, jd)$ is non-zero over many superlattice periods in this regime. An increase in d implies an increase in the barrier thickness for holes and electrons. As a result the total electron–hole bandwidth given by $4W(1)$ decreases, and the binding energy increases. For $d > 140$ Å, $W(1)$ is zero; hence the electron and hole are localised in the same layer. The superlattice exciton binding energy becomes equal to the value for a single quantum well which gradually decreases as d increases [13].

The presence of a finite electric field F along the growth direction adds

$$-eF\sum_i z_i$$

to the total exciton Hamiltonian (3). A term $eFjd$ consequently appears in the CCR exciton equation, and corresponds to *intra-sub-band* transitions induced by the electric field. Contributions arising from *inter-sub-band* transitions are neglected. The effect of the electric field on the zero-wavevector exciton energy spectrum is then described in the present theory by

$$\sum_{j,m}\left[\left(\varepsilon_j^{0,m} - eFjd - \varepsilon + \sum_{n=1}^{\infty} 2W(n)\right)\delta_{jj'}\delta_{mm'} - \sum_{n=1}^{\infty}\tilde{W}_{j'j}^{m'm}(n)\delta_{j'j\pm n}\right]c_j^m = 0. \tag{12}$$

In the zero-field limit, (12) is identical to (11). In the absence of the Coulomb interaction, $\varepsilon_j^{0,m} = 0$ and (12) leads to the formation of a Stark ladder [14].

Figure 2 compares the experimental [2] and theoretical exciton binding energies for the 40 Å GaAs/40 Å $Ga_{0.65}Al_{0.35}As$ superlattice as a function of increasing electric field F. The heavy-hole (C1–HH1) and light-hole (C1–LH1) binding energies shown correspond to the *intra-well* exciton, which is characterised by the electron and hole becoming localised in the same well at high electric fields. The total bandwidth of the electron and heavy hole ($4W(1)$) is 15 meV while that of the electron and light hole is 28 meV. At low fields the exciton binding energy tends to the zero-field result (cf figure 1). As F increases the binding energy increases due to the enhanced electron and hole localisation associated with the formation of the Stark ladder [14]. At high fields ($eFd \gg 4W(1)$) the exciton binding energies become those of an isolated quantum well of width 40 Å. The reason that the theoretical binding energies lie below the experimental values at high fields is most likely due to the neglect of *inter-sub-band* transitions and tunnelling effects.

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