Microscopic theory of Wannier-Mott excitons bound to monolayer insertions: the InAs in GaAs case

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Microscopic Theory of Wannier-Mott Excitons Bound to Monolayer Insertions: the InAs in GaAs Case

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A microscopic theory of Wannier-Mott excitons bound to monolayer insertions in semiconductors is presented. It is based on Green's function tight-binding calculations of the single-particle states. For one and two monolayers of InAs in GaAs, the binding energy and the oscillator strength are found to be large and to increase with the number of InAs layers.

An impurity plane in a bulk host matrix (like, e.g, InAs in GaAs or GaAs in AlGaAs) has a short-range potential which induces localization of electron and hole states and, consequently, of the exciton. This gives rise to observed strong luminescence in these systems, which can be thus useful in opto-electronic device applications [1 to 3]. A theoretical description of excitons bound to these ultra-narrow layers is required. From a fundamental point of view, monolayer (and submonolayer) impurity planes represent the ultimate limit of narrow quantum wells (QWs), with new and exciting theoretical problems due to the inadequacy of the usual envelope function (EF) scheme.

We present here a theoretical approach to excitons bound to monolayer (ML) impurities, which is based on the Green's function tight-binding (TB) scheme of [4,5]. We exploit the fact that the exciton is still Wannier-Mott like, as far as the in-plane dynamics is concerned. The relative electron–hole radius in the plane is, in fact, much larger than the lattice constant (and the in-plane exciton envelope function is strongly localized in k-space), thereby permitting a shallow-exciton approximation *for the in-plane motion only*. The single-particle dynamics is fully treated by the TB method. This mixed approach leads to a convenient way for calculating localized exciton states, without uncontrolled approximations in the physical description. We finally apply the method to the case of one and two MLs of InAs in GaAs, and the results are compared with those of the phenomenological EF model.

Expanding the exciton wavefunction in a basis set of Slater determinants [6], the k-space equation for the envelope function $A_{\mathrm{cv}}(\mathbf{k}_\mathrm{c}, \mathbf{k}_\mathrm{v})$ takes the form

$$[E_\mathrm{g} + \varepsilon_\mathrm{c}(\mathbf{k}_\mathrm{c}) - \varepsilon_\mathrm{v}(\mathbf{k}_\mathrm{v}) - E]\, A_{\mathrm{cv}}(\mathbf{k}_\mathrm{c}, \mathbf{k}_\mathrm{v})$$
$$-\sum_{\mathrm{c}',\mathrm{v}'} \sum_{\mathbf{k}'_\mathrm{c}, \mathbf{k}'_\mathrm{v}} \langle \psi_{\mathrm{c}'\mathbf{k}'_\mathrm{c}} \psi_{\mathrm{v}\mathbf{k}_\mathrm{v}} | \, \frac{e^2}{\varepsilon |\mathbf{r}_1 - \mathbf{r}_2|} \, | \psi_{\mathrm{c}\mathbf{k}_\mathrm{c}} \psi_{\mathrm{v}'\mathbf{k}'_\mathrm{v}} \rangle A_{\mathrm{c}'\mathrm{v}'}(\mathbf{k}'_\mathrm{c}, \mathbf{k}'_\mathrm{v}) = 0 \,, \qquad (1)$$

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where $\mathbf{k}_\mathrm{c}, \mathbf{k}_\mathrm{v}$ are two-dimensional (2D) Bloch vectors for the crystal with the impurity plane, c(v) is a conduction (valence) band index which may have a discrete and a continuous part, $\varepsilon_\mathrm{c}(\mathbf{k}_\mathrm{c})$ ($\varepsilon_\mathrm{v}(\mathbf{k}_\mathrm{v})$) is the conduction (valence) band dispersion. The electron–hole attraction is screened by the background dielectric constant ε, and the electron–hole exchange interaction is omitted. Writing the Bloch functions in terms of their periodic parts $u_{\mathrm{c},\mathbf{k}}$, $u_{\mathrm{v},\mathbf{k}}$, the Coulombic matrix element can be evaluated by expanding the products of periodic parts in plane waves with reciprocal lattice vectors $\mathbf{G}$, i.e.,

$$u^*_{\mathrm{c},\mathbf{k}_\mathrm{c}}(\mathbf{r})\, u_{\mathrm{c}',\mathbf{k}'_\mathrm{c}}(\mathbf{r}) = \sum_{\mathbf{G}} C(c\mathbf{k}_\mathrm{c}, c'\mathbf{k}'_\mathrm{c}; \mathbf{G})\, \mathrm{e}^{i\mathbf{G}\cdot\mathbf{r}}\,, \tag{2}$$

and similarly for the valence Bloch functions.

We consider only optically active exciton states, i.e., with $\mathbf{k}_\mathrm{c} = \mathbf{k}_\mathrm{v} \equiv \mathbf{k}$. The shallow-exciton approximation for the in-plane dynamics is defined by keeping only the $\mathbf{G} = \mathbf{0}$ reciprocal lattice vector in Eq. (2), and by neglecting the (small) variation of $u_{\mathrm{c},\mathbf{k}}, u_{\mathrm{v},\mathbf{k}}$, when the 2D wavevector varies on the scale of the inverse exciton radius. In this way we obtain a k-space equation similar to that of [7,8], but where the TB wavefunctions appear instead of the EF ones.

The wavefunctions are calculated in the TB Green's function formalism. The host crystal is described by a TB Hamiltonian H^0 in the basis of $\mathrm{sp^3s^*}$ atomic-like orthogonal orbitals centered on each atomic site [9]. Spin and strain have been consistently included [5]. In order to exploit the short-range nature of the isoelectronic impurity layer perpendicular to the layer plane, we represent the retarded Green's function matrix G^0 of the host material in a basis which is Bloch-like in the plane parallel to the impurity layer and Wannier-like perpendicular to it [10]. The perturbation induced by the impurity layer is then constructed according to the prescriptions of [4,5]. Measuring energies from the valence band top of GaAs, the heavy hole (HH1) and electron (CB1) confined levels are at 23 and 1468 meV for 1 ML, respectively, and at 54 and 1406 meV for 2MLs. The bound levels are close to the band edges of GaAs (whose gap is 1.52 eV), but far from those of InAs: this shows that an EF description of the impurity layer is unjustified.

The indexes c,v in Eq.(1) run over the discrete (bound) and continuum (unbound) states of the ML impurity plane. In the strong confinement regime, when there is separate quantization of electron and hole levels, the exciton wavefunction is well represented by keeping only the electronic states bound to the ML. Since the linear-chain TB model of [1], as well as the EF calculations of [11], indicate that the projection of the exciton onto the separable wavefunction, obtained by keeping only the bound state, is as high as 99% for 1 ML InAs in GaAs, we can make the approximation of keeping only the bound conduction and valence states in Eq. (1).

For optically active, s-state excitons, the envelope function $A(\mathbf{k})$ depends only on $k = |\mathbf{k}|$. The integral equations (1) are solved by the modified quadrature method described in [7,8]: the wavevector variable is discretized according to a Gaussian quadrature formula, while a singular function is added and subtracted in order to remove the divergence of the Coulomb potential, and the resulting matrix is diagonalized numerically.

Once the envelope function is obtained, the oscillator strength is calculated in the shallow-exciton approximation as

$$f_\varepsilon = \frac{2}{m_0 \hbar\omega} \left| \sum_{\mathrm{cv}} \langle u_{\mathrm{c},\mathbf{0}} |\, \varepsilon\cdot\mathbf{p}\, | u_{v,\mathbf{0}} \rangle \sum_{\mathbf{k}} A_{\mathrm{cv}}(\mathbf{k}) \right|^2 . \tag{3}$$

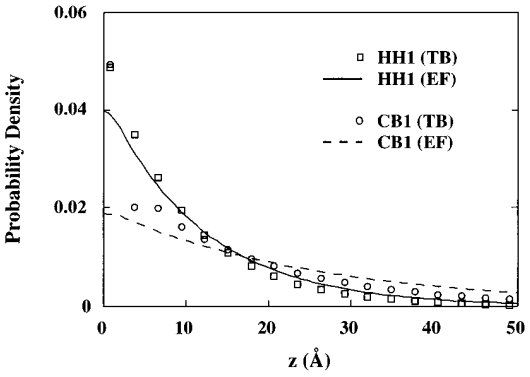

Fig. 1. Electron and hole probability densities along the growth direction for 1 ML InAs in GaAs. Circles/squares: tight-binding calculation, full/dashed lines: envelope function scheme

The momentum matrix element between Bloch functions of the impurity layer is expressed as the product of momentum matrix element between bulk Bloch functions times an electron–hole overlap integral.

The empirical TB scheme yields the planar average of Bloch functions for each atomic layer. A further average in the growth direction has been performed in order to eliminate the oscillations between anionic and cationic layers. These result is the analog of the macroscopic average used in band-offset calculations [12]. These average TB wavefunctions are compared to the result of the usual EF scheme for 1 ML of InAs in GaAs (see Fig. 1). The TB wavefunction of the electron is found to be more localized than its EF counterpart. The heavy-hole wavefunction is, instead, similar in the two methods.

In Table 1 we present our results for exciton binding energies and oscillator strengths for one and two MLs of InAs in GaAs. The calculations have been performed both in EF and TB approach. The present method allows to use the full in-plane dispersion calculated by the TB, without making any effective-mass approximation: however the in-plane dispersion might depend on the TB parametrization of the two bulk materials, which has to be carefully checked. Here we show preliminary results obtained assuming a parabolic in-plane dispersion with the GaAs mass. The effect of in-plane dispersion with the full TB results is being studied and will be published later on [13]. Binding energies and oscillator strengths are found to increase with the number of layer insertions, due to the increased localization of electron and hole wavefunctions. Moreover, the binding energies and oscillator strengths calculated with the present approach are always larger than the EF ones, due to the greater localization of the single-particle wavefunctions in the TB approach.

Table 1

Binding energies (in meV) and oscillator strenghts ($10^{12}\,\mathrm{cm}^{-2}$) for one and two MLs of InAs in GaAs, using in-plane masses of GaAs (see text).

	E_b		f/S	
	1 ML	2 ML	1 ML	2 ML
EF	8.1	9.7	4.5	6.6
TB	9.4	10.3	6.5	8.3

In conclusion, we have developed a microscopic approach to the study of the exciton bound to monolayer insertions. It takes into account the atomic-scale properties of the compounds in the heterostructures using the TB Green's functions model for the single-particle states, while keeping a Wannier-Mott approximation for the exciton in-plane dynamics.

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