Electronic structure and electron distribution in an inverse superatom calculated by a self-consistent method

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A full self-consistent procedure, applied to an inverse superatom structure is described. It is shown, both numerically and theoretically, that the electron concentration is large, but not maximal, the point of maximum being displaced off centre due to the fact that the second excited level has three times as many electrons as the ground level. Such an effect does not occur in classical quantum wells and superlattices. Moreover it is shown that the self-consistent treatment is necessary for an exact analysis of the energy band structure of the inverse superatom: solving the problem only by a trial rectangular potential gives an error of about 20%.

1. Introduction

Recent innovations in microfabrication technology have made it possible to make various semiconductor microstructures that are now opening the door to a new realm of physics called mesoscopic physics [1]. The simplest of them, the two-dimensional quantum wells and superlattices, have been very thoroughly studied, but one-dimensional structures (quantum wires) and zero-dimensional ones [2] have only recently attracted considerable attention. A number of quasi-zero-dimensional quantum structures have been proposed, the best known among them being the superatom, quantum dot, and semiconductor microcrystallites embedded in a glass matrix [3].

In this paper, we shall analyse the electronics of an inverse superatom or quantum dot (QD) self-consistently, and find conditions where such a calculation is necessary, and where its zeroth-order approximation, as we have used in [4], is accurate enough.

2. Outline of the calculation

For sphere (quantum dot) radii of the order of nanometres, the band-structure, potential distribution, and other relevant structure parameters should be calculated by a self-consistent solution of the Schrödinger and Poisson equations. Here we consider an undoped QD embedded in uniformly doped bulk (with donor

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concentration N_D). Holes may now be neglected, and an electron moves in an effective potential:

$$U(r) = -e\phi(r) + U_h(r) + U_{xc}(r) + U_{im}(r) \tag{1}$$

where $\phi(r)$ is the electrostatic potential, $U_h(r)$ the effective potential energy associated with the heterojunction discontinuity, $U_{xc}(r)$ the local exchange-correlation energy, and $U_{im}(r)$ the image potential (we neglect the image effects in U(r), which should be a good approximation for GaAs QD in $Al_xGa_{1-x}As$ bulk, due to the close values of their dielectric constants). Because of spherical symmetry, the envelope function for an electron in level (i,l) is to be found from the Schrödinger equation of the form:

$$-\frac{\hbar^2}{2m^*}\frac{d^2\chi}{dr^2} + \left[U(r) + l(l+1)\frac{\hbar^2}{2m^*r^2}\right]\chi = E\chi, \tag{2}$$

applied in segments $(0, R_0)$ and $(R_0, +\infty)$ separately, with the corresponding values of $m^* = m_1$ inside QD, and $m^* = m_2$ outside. The boundary conditions at the interface $r = R_0$ are the continuity of χ and $(1/m^*)\cdot d(\chi/r)/dr$. In eqn. (2), l is the azimuthal quantum number ($l = 0,1,2, \ldots$). The electron concentration is given by

$$n(r) = \frac{1}{2\pi}\sum_{l=0}^{l_{max}}(2l+1)\sum_{i=1}^{i_{max}(l)}\frac{\chi_{i,e}^2}{r^2}f_{FD}\left(\frac{E_{i,l}-E_F}{kT}\right)$$

$$f_{FD}(x) = [1 + \exp(x)]^{-1} \tag{3}$$

where E_F is the Fermi level. The summation over l in eqn. (3) is up to $l = l_{max}$, beyond which bound levels cease to exist, and over i ($i = 1,2,3, \ldots$) up to the largest (radial) quantum number of a bound state for any specific l.

The Poisson equation now takes the form:

$$\frac{1}{r^2}\frac{d}{dr}\left[\varepsilon(r)r^2\frac{d\phi}{dr}\right] = e[n(r)-N_D(r)],\ N_D(r) = 0,\ r < R_0 \text{ and}$$

$$N_D(r) = N_D,\ r > R_0 \tag{4}$$

with boundary conditions $\phi(0) = 0$ and $d\phi/dr(r=0)=0$. As the majority of electrons are confined to QD, there is a depleted volume in the bulk. Within the total depletion approximation, it has a finite width W, the neutrality holding for $r \geqslant R_0+W$, therefore $(d\phi/dr)_{W+R_0} = 0$. Due to the global neutrality of the structure we have:

$$\int_0^{W+R_0} n(r)4\pi r^2 dr = \int_{R_0}^{W+R_0} N_D \cdot 4\pi v^2 dv \Rightarrow \frac{1}{2\pi} \sum_{l=0}^{l_{max}} (2l+1) \sum_{i=0}^{i_{max}(l)} f_{FD}\left(\frac{E_{i,l}-E_F}{kT}\right) =$$

$$\frac{(W+R_0)^3 - R_0^3}{3} N_D \tag{5}$$

Moreover, the difference $[-e\phi(W+R_0)+U_h(W+R_0)]-E_F$ should be equal to $E_{FB} = E_{CB} - E_F$ in the bulk (measured from the conduction band edge in the bulk), determined from $n_{bulk}=N_D$. This is expressed by a rather lengthy equation, obtained from eqn. (4) in a straightforward manner, of the form:

$$\frac{\varepsilon_2-\varepsilon_1}{\varepsilon_1 \varepsilon_2} \frac{e^2}{\varepsilon_0(W+R_0)} \int_0^{R_0} n(v)\,(W+R_0-v)v dv - \frac{e^2}{\varepsilon_2\varepsilon_0} \int_0^{W+R_0} n(v) v dv +$$

$$+ \frac{e^2 N_D}{2\varepsilon_2\varepsilon_0} [(W+R_0)^2 - R_0^2] + U_0 = E_F + E_{FB} \tag{6}$$

where ε_1 and ε_2 are dielectric constants in dot and bulk, respectively. Equation (6) together with eqn. (5) enables one to determine W and E_F. Subsequently, n(r), ϕ(r) and $-e\phi + U_h$ are calculated (note that $U_h(r) = 0$ for $r<R_0$ and is equal to the band edge discontinuity, U_0, for $r>R_0$).

As for the exchange-correlation part, $U_{xc}(r)$, we use an analytic expression of the local-density-functional potential by Ceperley and Alder obtained by a Monte Carlo method and have introduced m* and ε in a way that had been done in a previous paper [2]. Afterwards this potential was added to the potential $-e\phi + U_h$ to obtain the potential U(r). We have initiated a self-consistent procedure by solving the Schrödinger equation (2) into which is fitted a trial potential having the form: $U_{trial}(r) = U_h(r)$, which means that $n(r) \equiv 0$ in the well and $n(r) = N_D$ in the bulk. Substituting condition $n(r) \equiv 0$ in eqn. (6) we arrive at $U_0 = E_F + E_{FB}$ and W=0. The trial potential is depicted by broken lines in Figs. 1 and 2.

3. Numerical results and discussion

Firstly, we present numerical results for a single example of a GaAs QD of radius $R_0 = 100$ Å in $Al_{0.35}Ga_{0.65}As$ bulk doped to $N_D = 10^{15}$ cm^{-3} at T = 77 K. This is a typical example with respect to structure parameters, and low N_D was chosen for the purpose of demonstrating the necessity for the self-consistent solution. In this case $V_0 = 270$ meV. As depicted in Fig. 1, the self-consistent U(r) departs considerably from the trial rectangular one, with band bending $\simeq$ 60 meV and a maximal field of $\simeq 10^7$ V m^{-1}. Furthermore, seven bound levels were found with U_{trial}, and only four remained upon completion of the self-consistent procedure. It is interesting to note that the point of electron density distribution is displayed off centre, due to the fact

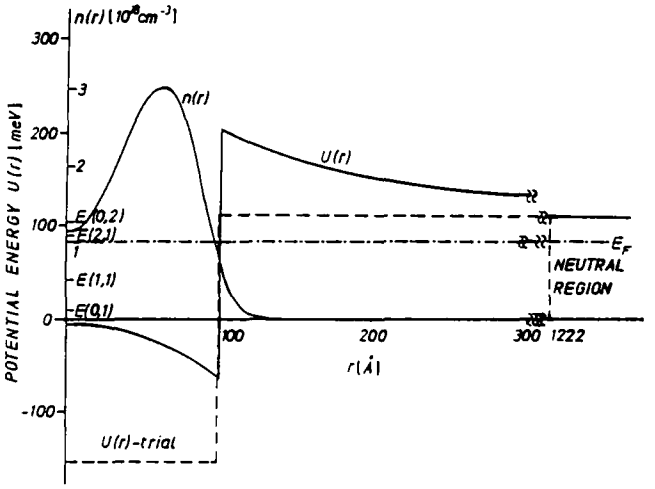

Fig. 1 The potential U(r) and electron concentration n(r) in a GaAs QD embedded in $Al_{0.35}Ga_{0.65}As$ bulk doped to $N_D = 10^{15}$ cm^{-3} (T=77 K). The trial, rectangular dependence, $U_{trial}(r)$, is given in broken lines

that E_F is above levels (l=0, i=1) and (l=1, i=1), making them almost fully populated, although the former is non-degenerate and the latter triply degenerate. For this same (degeneracy) reason, more electrons may be found on the higher (1,1) level than on the lower-lying (0,1) level under equilibrium conditions.

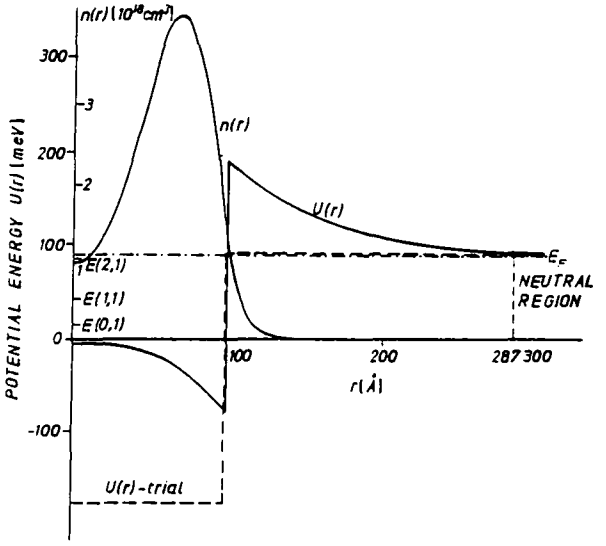

Fig. 2 Same as Fig. 1 except that $N_D = 10^{17}$ cm^{-3}

Figure 2 shows numerical results for the structure described above, but only for a high donor concentration $N_D = 10^{17}$ cm^{-3}. Furthermore, seven bound levels were found with U_{trial}, and only three remained upon the completion of the self-

consistent treatment. Fractional level occupations are 0.136, 0.409 and 0.453 for ground, first and second levels, respectively. The first and second levels are three- and five-fold degenerate, respectively. As the level E(2,1) is only 2 meV under the lowest continuous level, and therefore the fractional (2,1) level occupation is considerable (~ 50%), it could be assumed that influence of continuous levels on electron concentration will be significant, so that the model given in this paper should be expanded (this very complex problem is in the phase of final analysis and will be published elsewhere). It is interesting to note that electron concentration in a QD centre is 0.25–0.4 of the maximal concentration and originates from electrons at the ground level (l = 0). Electron concentration vanishes in the depletion region at a distance of about 30 Å from the interface. Influence of $U_{xc}(r)$ on the total distribution of potential is not great, as was expected, and is at its greatest at about 5 meV.

An important quantity in determining the absorption is certainly the difference between energy levels, as this determines directly the photon energy which corresponds to maximal absorption [4]. The difference mentioned has been defined as the difference between energy level E(l,i) and ground energy level E(0,1).

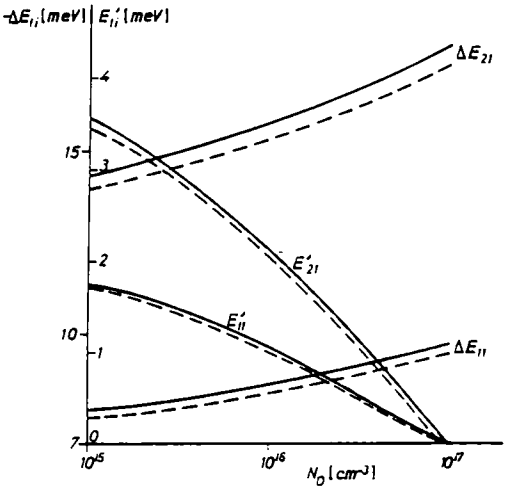

Fig. 3 The dependences ΔE_{li} and E_{li} vs. donor concentration N_D · ΔE_{li} is defined as $\{E(l,i)-E(0,1)\}^{\text{self-consistent}} - \{E(l,i)-E(0,1)\}^{\text{trial}}$, while $E_{li} = E_{li}(N_D)-E_{li}(N_D = 10^{17}\ cm^{-3})$ and $E_{li} = E(l,i)-E(0,1)$. The broken curves give the values calculated without inclusion of exchange and correlation effects. For $N_D = 10^{17}\ cm^{-3}$ the corresponding values of E_{21} and E_{11} are 75.64 (76.11) meV and 31.94 (32.15) meV with (without) inclusion of exchange and correlation effects, respectively

As a criterion for an appropriate self-consistent treatment we have introduced the difference $\Delta E_{li} = E_{li}^{\text{self-consistent}} - E_{li}^{\text{trial}}$. As shown in Fig. 3, the difference ΔE_{li} is always negative and considerable. For all values of donor concentration N_D from $10^{15}\ cm^{-3}$ to $10^{17}\ cm^{-3}$, ΔE_{11} and ΔE_{21} are in the ranges 7 - 10 meV and 14 - 18 meV, respectively. The importance of these differences can be illustrated by the following example. Let us assume that the bulk is doped to a concentration of $10^{17}\ cm^{-3}$. There is maximal absorption in the case of levels (0,1) and (2,1) at the photon wavelength of 16.4 μm, obtained from a complete self-consistent treatment, while the trial solution gives 13.25 μm, i.e. about 20% less. But knowing that linewidth is several meV,

the trial solution gives a neglected absorption at 16.4 μm. As ΔE_{1i} weakly depends on donor concentration (Fig. 3) a similar conclusion could be derived for $N_D = 10^{15}$ cm^{-3}; from this could be deduced the fact that the self-consistent treatment should be used for all donor concentrations of a practical nature. Finally, in Fig. 3 is shown dependence of E_{1i} on donor concentration. That dependence is very weak, i.e. E_{21} is reduced only ~ 4% if the donor concentration is increased a hundred times. Exchange-correlation potential, $U_{xc}(r)$, exerts very weak influences on ΔE_{1i} and E_{1i} of about 0.5 meV and 0.1 meV, respectively.

4. Conclusion

The self-consistent procedure of electronic structure calculations in inverse superatom (quantum dot) is described. The treatment is complete and based on effective mass and envelope function approximations. We have only neglected the influence of image potential because of the close agreement of the dielectric constants. Numerical results are presented for GaAs quantum dot of radius 100 Å in $Al_{0.35}Ga_{0.65}As$ bulk at T = 77 K. By varying the donor concentration from 10^{15} cm^{-3} to 10^{17} cm^{-3} the necessity of the self-consistent treatment has been demonstrated; i.e. if the photon wavelength which corresponds to maximal absorption is considered, the trial solution gives a wavelength about 20% lower. As the chosen example is characteristic, we can say that with small corrections it will be similar in other examples. This results in a general conclusion: the self-consistent treatment may be necessary for quantum dot-photodetector design.

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6. References

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