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LETTER TO THE EDITOR

A study of hydrogenic and metastable states of shallow-donor impurities in GaAs/GaAlAs multi-quantum wells

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Abstract. The energies of the excited states of a donor impurity in GaAs/GaAlAs multi-quantum wells are calculated using a model which is principally three-dimensional modified by the square well potential. Landau-like basis states are chosen and the effects of the Coulomb potential included using the techniques discussed by Simola and Virtamo. The results are found to be in very good agreement with our earlier calculations, using an alternative method, for those states which are hydrogen-like in weak fields. However, additional states are also obtained that are not hydrogenic in nature. These states are commonly termed 'metastable', in that they only exist in strong fields. The predictions of the model are shown to be in good agreement with experimental data obtained previously in photoconductivity experiments.

The problem of donor impurities that are confined in multi-quantum-well (MQW) heterostructures has attracted considerable attention in recent years. The donor energy levels can be probed experimentally by far-infrared (FIR) and inter sub-band spectroscopy. In addition, various theoretical techniques (e.g. perturbation theory, variational calculations) have been developed in order to understand the nature and properties of the impurity. More recently, Dellow *et al* [1] have shown that shallow donor impurities in a QW can give rise to additional features in the current–voltage characteristics of double-barrier resonant tunnelling structures due to the presence of donor-assisted tunnelling transitions. It is very clear, therefore, that it is necessary to provide an accurate quantum-mechanical description of the donor impurity in quantum-well devices.

The topic of donor and acceptor impurities in QW structures has been the subject of numerous experimental and theoretical papers within the last few months. We mention in particular the work of Mueller *et al* [2] on donor-level crossing in tipped fields, Radha Ranganathan *et al* [3] on the coupling between doped double-QW structures, Tsonchev and Goodfriend [4] for theoretical work on QW devices and also a theoretical study of the magnetopolaron bound to an impurity by Bao-Hua Wei and Shi-Wei Gu [5]. In addition, Shi *et al* [6], Hao Chen *et al* [7] and Weiquan Chen and Andersson [8] have used variational-type calculations in their respective modelling of differing properties of donor impurities in single and multiple QWs.

The work to be described here was motivated by the FIR photoconductivity experiments recently performed on silicon-doped GaAs/GaAlAs MQWs in Nottingham [9] in which several transitions were observed from the bound 1s-like ground state of the isolated impurity to various excited states in an applied magnetic field of up to 10 T.

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While some of the transitions observed were to the well-documented $2p_{+1}$- and $2p_{-1}$-like excited states, other transitions were observed for which no prior identification had been agreed. A theoretical model that allows transitions to some additional hydrogen-like excited states to be identified in the data was reported previously by us [10]. However, there are some additional experimental points that cannot be fitted by this theory. We believe that these arise from transitions to so-called metastable states, which have no hydrogenic counterpart and do not exist in the low-field limit. Previously [10], we tentatively identified some metastable states in FIR photoconductivity data, but were unable to predict their precise energies or obtain expressions for their wavefunctions.

Transitions to such metastable states have been observed in bulk GaAs by several groups of workers. The concept was first introduced for GaAs by Wagner and Prettl [11] in order to account for the magnetic field dependence of the photoconductivity experiments. Similar conclusions were made independently by Golubev *et al* [12]. The same theme was taken up by Klaassen and co-workers [13–16] who give a schematic energy level diagram for the hydrogen-like and metastable states and their relation to the Landau levels.

To our knowledge, however, no identification of metastable states in the MQW problem has been made by any other workers. Indeed, there has recently been some debate in the literature over whether metastable states should exist in MQWs. The basic question is whether a donor in a MQW should be treated as principally a two-dimensional (2D) or a three-dimensional (3D) system. Metastable states can only exist in a principally 3D model, as they effectively have infinite energies on a 2D model.

Details of an analytical model for the two-dimensional problem were given by MacDonald and Ritchie [17]. This 2D model was later used by McCombe and co-workers [18–20], and an alternative model developed by Larsen [21]. Larsen consequently concluded that there are no metastable resonant states in quantum well systems. While we agree with this statement in the case of a 2D atom, we believe that it is not appropriate for use in modelling the GaAs MQWs of width 150 Å or more, as investigated experimentally. For these cases, the well width is of at least the order of the effective Bohr radius (98.7 Å), and the wavefunction for the impurity electron is only slightly affected by the barrier confinement (see figure 4 of Dunn and Pearl [10], for example). Also, it seems more than coincidence that the graphs of transition energy against magnetic field are remarkably similar in the bulk and MQW cases (compare, for example, figure 2 of Wagner and Prettl [11] with figure 1 of Dunn *et al* [22]). As the well width decreases below 100 Å, 2D descriptions will become more appropriate and metastable states will effectively cease to exist.

In this letter, a theoretical model that allows the energies of metastable states in MQWs to be calculated will be developed. This approach is based on that described by Simola and Virtamo [23], who describe both hydrogenic and metastable states for hydrogen atoms in strong magnetic fields, such as those existing in stars, which is mathematically an equivalent situation to that in bulk GaAs. The method has been adapted here to model donor impurities in MQWs.

It is useful to refer to both hydrogenic and metastable states in strong magnetic fields using the high-field notation (N, m, ν), where N is the principal Landau quantum number, m is the usual magnetic quantum number and ν is the number of nodes of the wavefunction in the z direction. The ground 1s-like state is thus written as $(0, 0, 0)$. Transitions from this ground state must be to states $(N, 1, \nu)$ in order to satisfy the electric dipole selection rules (although other transitions may be allowed due to weak perturbative mixings). The observed sharpest transitions can be expected to be states with $\nu = 0$ in the above.

We begin with the non-relativistic Hamiltonian used by Simola and Virtamo [23] for the electron of a hydrogen atom in a magnetic field (neglecting spin), modified to take account of the effect of the MQW barriers. In this work we only consider the case of an impurity at the centre of a quantum well. Thus

$$\mathcal{H} = \boldsymbol{\pi}^2/2m + V(r) + V_B(z) \tag{1}$$

where $\boldsymbol{\pi} = \boldsymbol{p} + e\boldsymbol{A}$ is the momentum operator, $\boldsymbol{A}$ is the vector potential of the magnetic field and $V(r)$ is the Coulomb potential. $V_B(z)$ is the square well potential, taken to be 0 in the wells and a constant value V_0 in the barriers (60% of the band-gap difference $\Delta E_g = (1.155x + 0.37x^2)$ eV for $Ga_{1-x}Al_xAs$). The z axis is chosen to be perpendicular to the layers, which is also the direction of the uniform field $\boldsymbol{B}$.

It is useful to introduce operators

$$\pi_\pm = (\lambda/\hbar)(\pi_x \pm i\pi y)/\sqrt{2} \tag{2}$$

where $\lambda = \sqrt{\hbar/eB}$ is the magnetic length, which act as raising and lowering operators on N and m such that:

$$\pi_+ |N, m\rangle = \sqrt{N+1}\,|N+1, m+1\rangle \qquad \pi_- |N, m\rangle = \sqrt{N}|N-1, m-1\rangle. \tag{3}$$

Using these definitions and formulating the problem in dimensionless form, the Schrödinger equation becomes

$$[4\beta(\pi_+\pi_- + \tfrac{1}{2}) - d^2/dz^2 - 2/r + V_B(z)]\Psi = E\Psi. \tag{4}$$

The unit of length is the effecive Bohr radius, a_0 (98.7 Å for GaAs), and the unit of energy the effective Rydberg, R (5.83 meV for GaAs). β is a dimensionless measure of magnetic field:

$$\beta = e\hbar B/4mR \tag{5}$$

which is equal to 0.0735 B for GaAs, where B is in Tesla. In the adiabatic limit of infinitely strong magnetic fields, when the cylindrically-symmetric magnetic field dominates, the wave function $\Psi(r)$ for the electron can be written to a good approximation in the separated form

$$\Psi_{Nmv}(r) \sim \Phi_{Nm}(\rho, \varphi) f_m^N(z) \tag{6}$$

where Φ_{Nm} is the Landau wave function in cylindrical coordinates given by:

$$\Phi_{Nm}(\rho, \varphi) = (1/\sqrt{2\pi\lambda^2})\, e^{im\varphi}\, e^{-(1/2)\zeta}\, \zeta^{(1/2)|m|}\, P_{Nm}(\zeta) \tag{7}$$

where ζ is the dimensionless variable $\rho^2/2\lambda^2$.

The polynomials P_{Nm} are closely related to the associated Laguerre polynomials such that:

$$P_{Nm}(\zeta) = \frac{1}{\sqrt{N!s!}} \sum_{k=0}^{\min(N,s)} \frac{(-1)^k}{k!} \frac{N!}{(N-k)!} \frac{s!}{(s-k)!} \zeta^{\min(N,s)-k} \tag{8}$$

where $s \equiv N - m$. Substituting the separated form for Ψ and integrating over ρ and φ we thus obtain the differential equation

$$\left[-\frac{d^2}{dz^2} + 2\beta(2N+1) - 2\beta^{1/2} V_{NN}^m(\beta^{1/2}z) + V_B(z) \right] f_m^N(z) = E_{Nmv} f_m^N(z) \tag{9}$$

where

$$V_{NN'}^m(\beta^{1/2}z) = \int_0^\infty \frac{P_N^m(\zeta) P_{N'}^m(\zeta)\, e^{-\zeta}\, \zeta^{|m|}}{\sqrt{\zeta + (\beta^{1/2}z)^2}}\, d\zeta \tag{10}$$

(as $r^2 = \rho^2 + z^2$ for impurities at the centre of a quantum well). Note that the unit

of energy here is the effective Rydberg, R, rather than $2R$ as used by Simola and Virtamo [23].

This equation can be solved without difficulty using standard integration routines, and requiring ν nodes for a solution $\Psi_{Nm\nu}$. However, the results obtained, which strictly only apply to the adiabatic limit, are rather poor for the region of relatively low magnetic field accessible experimentally. For weaker fields, it is necessary to take into account the coupling between Landau levels due to the spherically-symmetric Coulomb potential. This can be done by writing the wave function $\Psi_{Nm\nu}$ in terms of basis states $\Phi_{N'm}$ in the form:

$$\Psi_{Nm\nu} = \sum_{N'=N_0}^{\infty} \Phi_{N'm} f_m^{N'}(z) \qquad \text{where } N_0 = \max(m, 0) \tag{11}$$

where $f_m^{N'}(z)$ are coefficients that are functions of z. This gives a system of coupled differential equations for $f_m^{N'}$ which can be solved numerically using an iterative procedure. However, this is a rather long process so is not attempted here.

In their analysis of the analogous bulk problem, Simola and Virtamo [23] found an asymptotic relation

$$f_m^{N'} = [V_{NN'}^m(\beta^{1/2}z)/2\beta^{1/2}(N' - N)] f_m^N \qquad (N' \neq N) \tag{12}$$

between the functions f_m^N and $f_m^{N'}$ which is appropriate for large but non-infinite fields. It seems reasonable to assume that a similar relationship will hold for the MQW case. Thus the differential equation (9) is modified to give

$$\left(-\frac{d^2}{dz^2} + 2\beta(2N+1) - 2\beta^{1/2} V_{NN}^m(\beta^{1/2}z) - \sum_{\substack{N'=N_0 \\ (N'\neq N)}}^{\infty} \frac{[V_{NN'}^m(\beta^{1/2}z)]^2}{(N'-N)} + V_B(z) \right) f_m^N$$
$$= E_{Nm\nu} f_m^N. \tag{13}$$

This is an eigenvalue problem which can be solved in the same manner as the adiabatic case, requiring the solution to have ν nodes for the state $\Psi_{Nm\nu}$. It is also found to be sufficient to restrict the upper limit on the sum to four terms in practice.

Figure 1 shows the transitions to the hydrogenic $2p_{-1}$ and $2p_{+1}$ states from the 1s ground state using the above method with the asymptotic correction. For comparison, it also shows the results obtained in our original hydrogenic method [10] (method 1). The calculations outlined in this paper are most accurate for high-energy excited states (Efros [24]), and are not particularly accurate for the 1s ground state. Consequently, the best picture obtainable is to take the ground state from our method 1 and the excited state from that presented here. It can be seen that, for fields above 10 T, there is only a small difference between the results. For very large fields, the difference between the results asymptotically approaches zero. This is somewhat surprising, as the hydrogenic method was not expected to be accurate for such large fields.

Figure 2 shows the calculated transition energies from the 1s state of method 1 to the hydrogenic $2p_{\pm}$ states from method 1 and the metastable $(1, -1, 0)$, $(2, 1, 0)$, $(3, 1, 0)$ and $(4, 1, 0)$ states from this method together with the experimental data of Grimes *et al* [9] and Cheng and McCombe [18]. The calculations are for wells and barriers that are 150 Å wide, and with an Al concentration of $x = 0.33$. The experimental data are for nominally similar values. It can be seen that the upper sets of data points lie very close to the predicted $(4, 1, 0)$ and $(3, 1, 0)$ metastable states. For the states identified as $(2, 1, 0)$ and $(1, -1, 0)$ the results are poorer, which is consistent with the observation

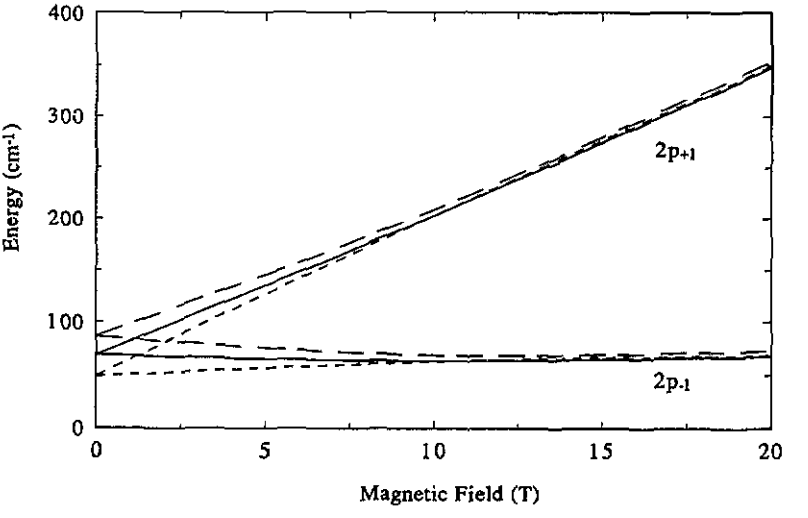

Figure 1. Transitions from the 1s ground state to the $2p_{\pm 1}$ excited states: the lines – – – are from the present method and the solid lines from method 1. The lines —— are obtained using the 1s state of method 1 and 2p states from the present method.

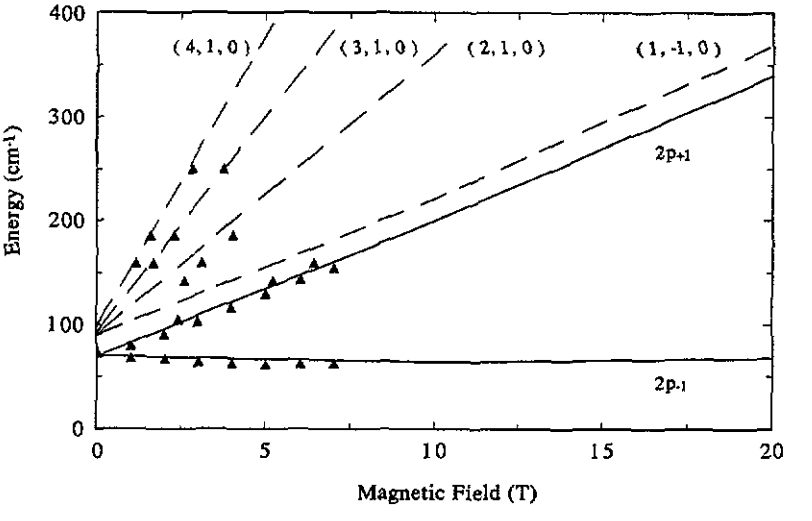

Figure 2. A comparison of the theoretical predictions with the experimental data [9, 18] (▲). All results were obtained using the 1s ground state of method 1. The solid lines are for the $2p_{\pm}$ states of method 1 and the broken lines various states from the present approach, labelled by the high-field notation (N, m, ν).

of Efros [24]. However, the agreement is still good considering that the method should only work for very strong fields. Transitions to all of these metastable states are expected to be strong as they satisfy the required $\Delta m = \pm 1$ selection rule. Such transitions are known to dominate the photoconductivity experiments on bulk GaAs [12–16].

This is the first time that a quantitative explanation of the data points at the higher energies has been given. The assignments agree with those obtained in Dunn and Pearl

[10] by a qualitative comparison of the bulk and MQW cases and energy separation arguments. It should be noted that the results of method 1 presented here are obtained taking an isotropic effective electron mass of 0.067 m_e throughout the sample, as this is the parameter used in this current work so far. In our original paper [10], different masses were introduced for the well and barrier materials, and also for the parallel and perpendicular directions (to simulate the effect of perturbations such as the polaron effect). This gives a much better agreement between the theoretical predictions and experimental results.

The method presented here is our first calculation of metastable states and their energies in MQW systems. In the future we hope to produce better results away from the adiabatic limit by iteratively solving the differential equations obtained using the states (11). We can also include variable masses and polaron effects, for example. This should greatly improve the agreement with lines such as (2, 1, 0) and (1, −1, 0), which cannot be obtained using the hydrogenic method and are also not accurate in the adiabatic limit of this method.

The agreement between the experimental points and the new theory clearly supports our treatment of the silicon donor in the GaAs well as being three dimensional. The calculations are significantly more complicated than those in two dimensions and give the important result that many more transitions satisfying the selection rules are allowed. It would appear from the results that metastable states do exist in MQWs as well as in bulk GaAs.

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