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On the stability of two-dimensional negative donor centres D^{2-} in high magnetic fields

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Abstract. Two-dimensional negatively charged donor centres D^{2-} (i.e. the states formed by a neutral donor D^0 binding two extra electrons) in a strong magnetic field with electrons confined to the spin-split zero Landau levels $0\downarrow$, $0\uparrow$ are considered. When the admixture of higher Landau levels is neglected, the energies and eigenfunctions of the doublet $S = \frac{1}{2}$ and quadruplet $S = \frac{3}{2}$ states are obtained. It is shown that both the doublet and the quadruplet D^{2-} states, although localized, lie above the ground D^- singlet and triplet states, respectively, i.e. have negative binding energies and, hence, are thermodynamically unstable against separation of an electron.

1. Introduction

Recently Huant *et al* [1,2] in a series of magneto-optical experiments have shown that by selective doping of $GaAs/Ga_{1-x}Al_xAs$ quantum wells (QWs) it is possible to 'engineer' a stable population of negative donor centres D^- (i.e. neutral donors trapping a second electron) or even to convert *all* neutral donors within a QW to D^- [3]. Since the work of Huant *et al* the problem of a quasi-two-dimensional (Q2D) D^- centre in a strong magnetic field, one of the simplest correlated quantum-confined systems, has attracted much attention.

Theoretical consideration of Q2D D^- centres in magnetic fields includes the quantum diffusion Monte Carlo studies of Pang and Louie [4] of the singlet D^- s ground state, the consideration of a strictly two-dimensional (2D) limit in a strong-magnetic-field approximation [5,6] where D^- eigenstates can be found exactly (see also [7]), variational calculations of the singlet D^- binding energies with the inclusion of the polaron corrections of Shi *et al* [8] and variational calculations of D^- singlet transition energies [9] for wide (510 Å) $GaAs/Ga_{1-x}Al_xAs$ QWs and fields $B < 6$ T (see also [10]). In particular, Larsen and McCann [5] and Dzyubenko [6] have independently shown that the strong magneto-optical transitions of the D^- singlet lead to final excited localized D^- states, and not a continuum (as was assumed in [1,4,8]) which in the relevant region of energies is absent. As a result, D^- transition energies exceed considerably the D^- binding energy and, hence, direct comparison between the theories [4,8] and magneto-optical experiments is impossible. More recently, direct

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diagonalization of the Hamiltonian for narrow (100 Å) $GaAs/Ga_{1-x}Al_xAs$ QWs with a consistent treatment of the non-parabolicity and subsequent inclusion of magneto-polaron corrections [11] enables one to derive the energies and line strengths of the dipole-allowed spin-singlet and spin-triplet magneto-optical transitions of D^-.

With increasing B the enhancement of attractive electron–impurity interactions exceeds the enhancement of electron–electron repulsion; thus B deepens the D^- singlet state and, just as in a 3D situation (see [12, 13], and references therein), makes several D^- triplet states stable [5–7]. In confined geometry and in the presence of quantizing magnetic fields, one can expect that even more electrons could be bound by a single donor ion, forming D^{2-} states with three bound electrons.

For a 2D case, Bychkov *et al* [7] have proposed a general method of constructing the quadruplet $S = \frac{3}{2}$ D^{2-} states in the zero Landau level and have calculated the energy of the non-degenerate $|M_z| = 3$ D^{2-} state (which, however, contains an error). Bychkov and Rashba [15] considered electron correlations in the zero $0\downarrow$, $0\uparrow$ levels in the presence of a donor ion for both $S = \frac{3}{2}$ and $S = \frac{1}{2}$ D^{2-} states (in [15], however, only the lowest two channels, s and p, both in electron–electron and electron–impurity interactions were taken into account). The present paper is an extension of the work of Bychkov *et al* [7]. Here we shall systematically study the spectra of D^{2-} states with electrons in the zero Landau levels $0\downarrow$, $0\uparrow$ and discuss the *stability* of D^{2-} states.

2. Eigenenergies and stability of D^{2-}

Following [7] (see also [14] and [15]) we obtain the exact D^{2-} eigenenergies and eigenstates from the secular equation which involves the matrix elements of the interaction Hamiltonian

$$H = -\sum_{i=1}^{3} \frac{e^2}{\epsilon r_i} + \frac{1}{2}\sum_{i\neq j}^{3} \frac{e^2}{\epsilon|\mathbf{r}_i - \mathbf{r}_j|} \equiv V_{\text{imp}} + H_{\text{ee}} \tag{1}$$

between non-interacting wavefunctions in the zero Landau levels which (omitting the exponential part) are polynomials in the complex variables $\bar{z}_j = x_j - iy_j$, $j = 1, 2, 3$. Admixture of higher levels can be neglected when $l_B = (\hbar c/eB)^{1/2} \ll a_B = \epsilon\hbar^2/m^*e^2$. If we change from the orthogonal transformation to the coordinates [7, 14]

$$Z = (z_1 + z_2 + z_3)/\sqrt{3} \qquad z_a = (z_1 + z_2 - 2z_3)/\sqrt{6} \qquad z_b = (z_1 - z_2)/\sqrt{2} \tag{2}$$

the complete orthonormal basis set of states with the total spin S, its projection S_z and with the total angular momentum projection $M_z = -(i + k + 2n)$ is given by

$$|i, k, n\rangle_{S,S_z} = a_i^{-1}(\bar{Z})^i |k, n\rangle_{S,S_z} \equiv |N\rangle \tag{3}$$

where $a_m = (2\pi l_B^2 2^m m!)^{1/2} l_B^m$ and $|k, n\rangle_{S,S_z}$ is the wavefunction of the relative motion (a uniform polynomial in the variables $\bar{z}_a$ and $\bar{z}_b$). For antisymmetric polynomials for the quadruplet $S = \frac{3}{2}$ states we shall use Laughlin's [14] representation, writing it in the form

$$|k, n\rangle_{3/2,S_z} = \sqrt{2}a_{k+n}^{-1} a_n^{-1}[(\bar{z}_1^k - \bar{z}_2^k)/2i](\bar{z}_1\bar{z}_2)^n S_{3/2,S_z} \tag{4}$$

where $\tilde{z}_{1(2)} = (\tilde{z}_a \pm i\tilde{z}_b)/\sqrt{2}$ and $S_{3/2,S_z}(\sigma_i)$ is the spin part of the wavefunction (e.g. $S_{3/2,-3/2} = |\downarrow\downarrow\downarrow\rangle$) and the quantum numbers in (4) take the values

$$k = 3m \qquad m = 1, 2, \ldots \qquad n = 0, 1, \ldots . \tag{5}$$

To obtain mathematically close descriptions for both $S = \frac{3}{2}$ and $S = \frac{1}{2}$ D^{2-} states, we extend Laughlin's representation (4) to the doublet $S = \frac{1}{2}$ states and construct the *total* antisymmetric wavefunction of the relative motion (which is convenient in computing the matrix elements; see (10) and (11) below). Choosing for a 2D basis (see, e.g. [16]) of the spin part of the wavefunctions the states (e.g. for $S_z = -\frac{1}{2}$)

$$S^{(1)}_{1/2,-1/2} = (1/\sqrt{6})\{|\downarrow\uparrow\downarrow\rangle + |\uparrow\downarrow\downarrow\rangle - 2|\downarrow\downarrow\uparrow\rangle\} \tag{6}$$

$$S^{(2)}_{1/2,-1/2} = (1/\sqrt{2})\{|\downarrow\uparrow\downarrow\rangle - |\uparrow\downarrow\downarrow\rangle\} \tag{7}$$

we find that

$$|k, n\rangle_{1/2,S_z} = F^{(1)}_{k,n}(\tilde{z}_a, \tilde{z}_b)S^{(1)}_{1/2,S_z} + F^{(2)}_{k,n}(\tilde{z}_a, \tilde{z}_b)S^{(2)}_{1/2,S_z} \tag{8}$$

where $F^{(1)}_{k,n}$, $F^{(2)}_{k,n}$ is a pair of two uniform polynomials given by (compare (4) and [15])

$$\left\{F^{(1)}_{k,n}, F^{(2)}_{k,n}\right\} = a^{-1}_{k+n}a^{-1}_n\{(\tilde{z}_1^k - \tilde{z}_2^k)/2i, \pm(\tilde{z}_1^k + \tilde{z}_2^k)/2\}(\tilde{z}_1\tilde{z}_2)^n. \tag{9}$$

Here $k = 3m + 1$ for the upper (+) sign and $k = 3m + 2$ for the lower (−) sign in (9); $m, n = 0, 1, 2, \ldots$.

The matrix elements of the Hamiltonian of electron–impurity interactions (V_{imp}) and of electron–electron interactions (H_{ee}) from (1) are evaluated in the variables $Z' = (z_1 + z_2)/\sqrt{2}$, z_b, z_3 and from (2) as

$$\langle N'|V_{\text{imp}}|N\rangle = -3\langle N'|e^2/\epsilon|z_3\|N\rangle \tag{10}$$

$$\langle N'|H_{\text{ee}}|N\rangle = (3/\sqrt{2})\langle N'|e^2/\epsilon|z_b\|N\rangle \tag{11}$$

respectively, and hence are reduced to the matrix elements of the electron–impurity interaction in the zero Landau level V_m:

$$V_m = [(2m-1)!!/2^m m!]E_0 \qquad E_0 = (\pi/2)^{1/2}(e^2/\epsilon l_B) \propto \sqrt{B}. \tag{12}$$

V_m give the binding energies of the ground $m = 0$ ($V_0 \equiv E_0$) and excited $m = 1, 2, \ldots$ states of a 2D neutral donor D^0 with an electron in the zero Landau level [17].

The calculated eigenenergies of the interaction Hamiltonian (1) for the doublet $S = \frac{1}{2}$ states with $|M_z| \leqslant 15$ are plotted in figure 1 and for the quadruplet states

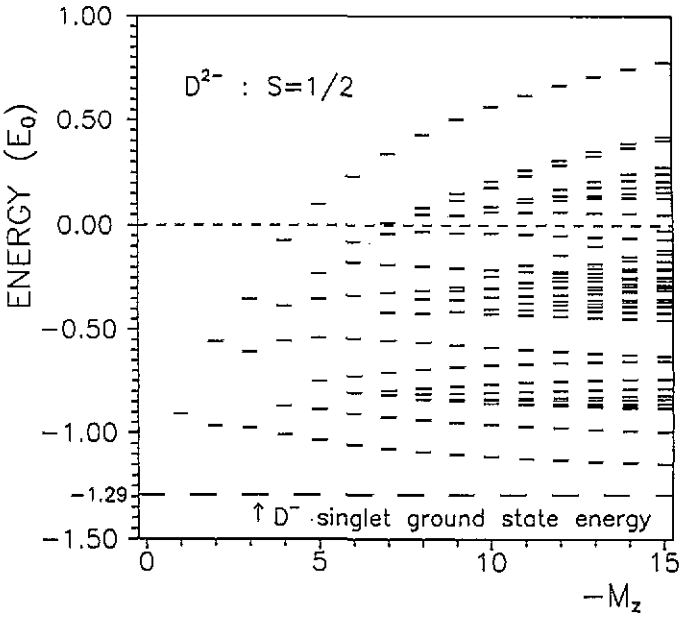

Figure 1. Interaction energies (in units of $E_0 = (\pi/2)^{1/2}(e^2/\epsilon l_B)$, the D^0 ground-state binding energy [17]) versus M_z for total spin of electrons $S = \frac{1}{2}$. All $D^{2-}_{S=1/2}$ states lie above the singlet ground state of D^- (shown by the broken line with long dashes) and have negative binding energies. The state with $M_z = 0$ is absent owing to the Pauli exclusion principle.

with $S = \frac{3}{2}$ in figure 2. The spin energies $g^*\mu_B B S_z$ of electrons in a magnetic field are not taken into account explicitly throughout this paper.

The excited branches of the spectra in which the energy is an increasing function of $|M_z|$ emerge when the quantum numbers k and n of the relative motion are fixed low. Physically, such states correspond to a situation when all three electrons in close proximity to each other are moved away from the donor ion D^+ (compare the discussion of the D^- spectra in [6]). The upper bounds of the spectra, asymptotically achieved at $|M_z| \gg 1$, are given by the largest energies in a system of three electrons (without D^+), $1.5910E_0$ for $S = \frac{1}{2}$ and $0.9612E_0$ for $S = \frac{3}{2}$, i.e. by the eigenenergies of the Hamiltonian H_{ee} in the states $|0,1,0\rangle_{1/2}$ and $|0,3,0\rangle_{3/2}$ from (11).

The lowest branches of the spectra with the energies decreasing with increasing $|M_z|$ correspond to a situation when two electrons are bound to the donor ion, forming D^- in the ground state, while the third electron is moved away from the donor complex to the maximal (allowed at the given M_z) distance. For the total spin $S = \frac{1}{2}$ of three electrons, D^- can be either in the triplet or in the singlet ground state; for $S = \frac{3}{2}$, only the triplet D^- ground state can be realized. Hence, the asymptotic values of these branches (shown by the broken lines with long dashes in figures 1 and 2) are given by the D^- singlet and triplet ground-state energies [5–7], $-1.2929E_0$ and $-1.1465E_0$, respectively.

Between these two limiting cases of the low-lying and excited branches there are also the following.

(i) A quasi-continuum spectrum should occur when all three electrons with

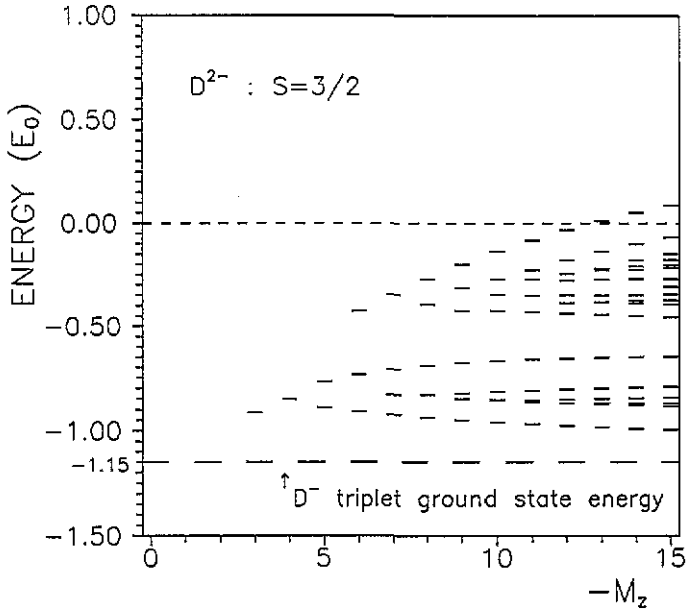

Figure 2. Interaction energies versus M_z for total spin of electrons $S = \frac{3}{2}$. All $D^{2-}_{S=3/2}$ states lie above the triplet ground state of D^- (shown by the broken line with long dashes) and have negative binding energies. The states with $M_z = 0, -1, -2$ are absent owing to the Pauli exclusion principle.

increasing $|M_z|$ are distributed over further larger areas, weakly interacting with each other and the donor ion D^+.

(ii) For rather large $|M_z|$ in the spectrum, states corresponding to the ground and excited states of D^0 whose energies are slightly shifted upwards owing to the presence of two electrons at large distances from a D^0 complex should appear.

The important point which we wish to stress is that there are no D^{2-} states with energies below firstly the D^- singlet ground state for $S = \frac{1}{2}$ and secondly the D^- triplet ground state for $S = \frac{3}{2}$, i.e. all D^{2-} states have negative binding energies. Therefore, D^{2-} states are *thermodynamically* unstable against the separation of an electron via

$$D^{2-}_{S=3/2(1/2)} \rightarrow D^{-}_{S=1(0)} + e^{-}. \tag{13}$$

However, it should be noted that, because of the localizing effect of a magnetic field B, all states of 2D electrons in the presence of a donor ion D^+ are discrete and, strictly speaking, there is no continuum. Therefore, an electron which goes to infinity from D^{2-} should pass through the intermediate states with larger radii having larger magnetic quantum numbers. (Obviously, at some step the isolated impurity picture ceases to be valid in that process. In other words, the broadening of Landau levels due to a finite impurity concentration n_{imp} (and other factors) should be taken into account. Here it is assumed that the impurity broadening of Landau levels is small compared with E_0. This is expected to be the case for impurity concentrations such that $2\pi l_B^2 n_{\text{imp}} \ll 1$.)

Hence, 2D D^{2-} states in high fields (at least in principle) can be metastable and (in a solid body) they become unstable at low temperatures through a cascade emission of acoustic phonons. The possibility of observing D^{2-} states in Q2D semiconductor QWs in high fields for non-equilibrium conditions (e.g. under photoexcitation) depends on their lifetimes which, through the level energy positions and acoustic-phonon-assisted transition matrix elements, are B dependent.

References

[1] Huant S, Najda S P and Etienne B 1990 *Phys. Rev. Lett.* **65** 1486
[2] Huant S, Mandray A, Martinez G, Grynberg M and Etienne B 1992 *Surf. Sci.* **263** 565
[3] Mandray A, Huant S and Etienne B 1992 *Europhys. Lett.* **20** 181
[4] Pang T and Louie S G 1990 *Phys. Rev. Lett.* **65** 1635
[5] Larsen D M and McCann S Y 1992 *Phys. Rev.* B **45** 3485
[6] Dzyubenko A B 1992 *Phys. Lett.* A **165** 357; 1992 *Fiz. Tverd. Tela* **34** at press (Engl. Transl. 1992 *Sov. Phys.–Solid State* **34** at press)
[7] Bychkov Yu A, Iordanskii S V and Eliashberg G M 1981 *Pis. Zh. Eksp. Teor. Fiz.* **33** 152 (Engl. Transl. 1981 *JETP Lett.* **33** 143)
[8] Shi J M, Peeters F M and Devreese J T 1993 *Physica* B at press
[9] Mueller E R, Larsen D M, Waldman J and Goodhue W D 1992 *Phys. Rev. Lett.* **68** 2204
[10] Larsen D M and McCann S Y 1992 *Phys. Rev.* B **46** 3966
[11] Dzyubenko A B and Sivachenko A Yu 1993 to be published
[12] Larsen D M 1979 *Phys. Rev. Lett.* **42** 742; 1979 *Phys. Rev.* B **20** 5217
[13] Najda S P, Armistead C J, Trager C and Stradling R A 1989 *Semicond. Sci. Technol.* **4** 439
[14] Laughlin R B 1983 *Phys. Rev.* B **27** 3383
[15] Bychkov Yu A and Rashba E I 1989 *Zh. Eksp. Teor. Fiz.* **96** 757 (Engl. Transl. *Sov. Phys.–JETP* **69** 430)
[16] Elliot J P and Dowber P G 1979 *Symmetry in Physics* vol 2 (London: Macmillan) ch 17
[17] Lerner I V and Lozovik Yu E 1980 *Zh. Eksp. Teor. Fiz.* **78** 1167 (Engl. Transl. *Sov. Phys.–JETP* **51** 588)