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Binding energies and photoionization transitions of two-dimensional negative donor centers D^- in high magnetic fields

Alexander B. Dzyubenko
Research Centre for Technological Lasers, Academy of Sciences of Russia, 142092 Troitsk, Moscow District, Russia

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The binding energies of two-dimensional (2D) negative donor centers D^- with two electrons bound to the donor ion in the singlet and triplet states in a strong magnetic field are obtained. It is shown that the energies of photoionization transitions $D^- \to N$, in which one electron is excited from zero to the Nth Landau level exceed considerably the binding energies of D^-. The absorption by the triplet ground state is shown to differ from that of the singlet by an additional line which occurs at energies *lower* than the cyclotron energy $\hbar\omega_c$ and the corresponding dipole-allowed transition does not lead to the photoionization of D^-.

1. Introduction

There is an increasing interest, both experimental and theoretical, in the magnetic field effect on shallow-impurity states in semiconductor quantum wells (QWs) and superlattices (see e.g. refs. [1–8]). Selective doping of a GaAs/GaAlAs QW makes it possible to realize the situation in which part of the electrons weakly bound to shallow donors in the GaAlAs barrier are transferred to the GaAs QW. In a QW they are trapped by neutral donors D^0 forming there a stable population of negatively charged donor centers D^- containing two electrons [9]. D^- states are most revealingly identified in strong magnetic fields H by the photoionization transitions $D^- \to N$ in which one electron is excited from zero to the Nth Landau level leaving behind a neutral D^0 [9,10]. The binding energy of D^-, E_b, experimentally determined [9] as the energy of the $D^- \to N=1$ photoionization transition diminished by the cyclotron energy $\hbar\omega_c = \hbar eH/m^*c$ is in agreement with E_b derived from the fundamental $D^- \to N=0$ photoionization transition and, also, with the quantum Monte Carlo calculations of the D^- singlet s-ground state [11] only when the polaron corrections [12] are taken into account.

In this Letter, considering a strictly 2D model in a high magnetic field first proposed in ref. [13] (see also ref. [14]), we intend to demonstrate that the energies of $D^- \to N$ photoionization transitions exceed considerably the binding energies of D^- ground states. The reason for this being that after the absorption of an infrared photon, D^- turns out to be in the final *excited* unstable state which requires additional energy compared to E_b.

It should be also noted that due to the low values of the effective g^*-factor of electrons in GaAs, transitions both from the singlet and the triplet states of D^- should be taken into account. 2D triplet D^- states, just as in a 3D situation (see refs. [15,16]) can be stable in a magnetic field. We will show that the absorption by the triplet p-ground state differs from that of the singlet s-ground state by an additional line associated with the strong dipole-allowed transition to a final stable D^- state.

2. Classification of states and D^- wave functions

In our consideration we shall neglect mixing between different Landau levels which is valid for magnetic fields H such that

$$\hbar\omega_c \gg E_0 = \left(\frac{\pi}{2}\right)^{1/2} \frac{e^2}{\epsilon r_H} \propto \sqrt{H}, \qquad (1)$$

where $r_H = (\hbar c/eH)^{1/2}$ and E_0 is the typical energy of Coulomb interactions in a strong magnetic field. (The magnetic field H corresponding to

$$\left(\frac{2}{\pi}\right)^{1/2} \frac{E_0}{\hbar\omega_c} = \frac{r_H}{a_B^*} = 1$$

for GaAs with the effective Bohr radius $a_B^* = 98.7$ Å and $Ry^* = 5.83$ meV is $H = 6.73$ T.)

Following ref. [13] (see also ref. [17]), we construct wave functions of 2D electron–impurity complexes out of the non-interacting wave functions. For a systematic treatment of the interaction matrix elements, it is convenient to use the factored wave functions of charged particles in a magnetic field [18–20],

$$\phi_{n,m}(\boldsymbol{r}) = \langle \boldsymbol{r} | nm \rangle = \frac{(a^+)^n (b^+)^m}{(2\pi r_H^2 n! m!)^{1/2}} \exp\left(-\frac{|z|^2}{4r_H^2}\right) \qquad (2)$$

(see refs. [19,20] for the explicit form of the ladder Bose operators a^+, b^+), $z \equiv x + iy$. In (2) n is the Landau level number, m, the oscillator quantum number, in which the energy is macroscopically degenerate. n, m are connected with the projection of the angular momentum m_z by the relation $m_z = n - m$ and determine the square of the orbit radius by $\langle r^2 \rangle_{nm} = 2(n+m+1) r_H^2$.

In a strong magnetic field the states of an electron–impurity complex can be characterized by the quantum numbers $\{S, S_z, N, M\}$. Here S and S_z are the total spin and its projection in the magnetic field direction, N is the *total* Landau level number, connected with the Landau level numbers of individual electrons n_i (non-conserved in the presence of interactions) as $\sum_i n_i = N$. (Note that it is at this point that we resort to the strong magnetic field approximation: we neglect virtual transitions to the states with $N' \neq N$, since they are small as powers of $E_0/\hbar\omega_c \ll 1$.) Physically, N describes the state in the Landau-fan-type diagram picture. M is the total oscillator quantum number $M = \sum_i m_i$ related to the projection of the total angular momentum as $M = N - M_z$. Since for the case of central potentials M_z is the *exact* quantum number and N is fixed, M is conserved.

In the effective mass approximation, the D^- eigenvalues and eigenfunctions are to be determined from the secular equation which involves the matrix elements of the interaction Hamiltonian

$$\hat{H} = -\frac{e^2}{\epsilon r_1} - \frac{e^2}{\epsilon r_2} + \frac{e^2}{\epsilon |\boldsymbol{r}_1 - \boldsymbol{r}_2|} \qquad (3)$$

between different wave functions with fixed N, M and S constructed out of (2). It is appropriate to pass by an orthogonal transformation to the coordinates [13] $\boldsymbol{r} = (\boldsymbol{r}_1 - \boldsymbol{r}_2)/\sqrt{2}$, $\boldsymbol{R} = (\boldsymbol{r}_1 + \boldsymbol{r}_2)/\sqrt{2}$ in which the complete orthonormal basis set with fixed N, M is given by

$$\phi_{n,m}(\boldsymbol{r}) \phi_{N-n,M-m}(\boldsymbol{R}) \qquad (4)$$

with different (integer non-negative) quantum numbers n, m. The basis set (4) includes a finite number of different states $\mathcal{N}$ – in contrast to the case of 2D magnetoexciton complexes [21], where $\mathcal{N} = \infty$. Since the parity of $\phi_{n,m}(\boldsymbol{r})$ is $(-1)^{n+m} = (-1)^{M_z}$, for the singlet $S = 0$ (triplet $S = 1$) states $n+m$ should be even (odd). It can be directly calculated that there are $\mathcal{N} = (N+1)(M+1)/2$ different states in the basis set (4), except in the case when both N and M are even; then $\mathcal{N} = (NM+N+M)/2+1$ for $S = 0$, and $\mathcal{N} = (NM+N+M)/2$ for $S = 1$.

In the calculations we restrict ourselves to the cases when (i) both electrons are in the zero Landau levels, i.e. $N = 0$ and (ii) one electron is excited to the first Landau level $N = 1$. In the representation chosen, when the interaction Hamiltonian (3) is expressed in the variables $\boldsymbol{r}$ and $\boldsymbol{R}$, the matrix elements of e–e interactions are diagonal and are expressed through the energies of the electron–impurity interactions for the $n = 0$ and $n = 1$ Landau levels,

$$V_{0,m} = \frac{(2m-1)!!}{2^m m!} E_0, \quad V_{1,m} = \frac{4m-1}{4m-2} V_{0,m}. \qquad (5)$$

Vice versa, the matrix elements of electron–impurity interactions turn out to be off-diagonal and are expressed through the matrix elements of pair-wise electron interactions in the zero Landau levels $U_{kl}^{(00)}(p)$ and the direct and exchange interactions in the zero and the first Landau levels, respectively, $U_{kl}^{(01)}(p)$ and $U_{kl}^{(01)E}(p)$. Explicit expressions will be presented elsewhere [22] (see also refs. [20,21]).

Note that for fixed k, l the matrix elements decrease exponentially with increasing p.

3. Results and discussion

The eigenenergies of the interaction Hamiltonian (3) for $M=N-M_z\leqslant 20$ for the singlet and triplet states in the zero Landau levels $N=0$ are depicted in figs. 1a, 1b and for $N=1$ in figs. 1c, 1d.

(1) Let us first discuss some general features of the D^- spectra. From figs. 1a–1d it is seen that with increasing M an increasing number of different "branches" are emerging. The low-lying "horizontal" branches physically correspond to the D^0 states in which one electron is bound to the donor ion with quantum numbers $m=0, 1, \ldots$, while the second electron moves away from D^0 to the maximal (allowed at the given M) distance. The asymptotical values of these branches are given by the electron–impurity interaction energies $\{-V_{0,m}\}$ for $N=0$ and $\{-V_{0,m}, -V_{1,m}\}$ for $N=1$ placed in increasing order. The first few values (in units of E_0) are $-V_{0,0}=-1$, $-V_{1,1}=-0.75$, $-V_{0,1}=-V_{1,0}=-0.5$ (and for $N=1$ there are *two* practically degenerate branches, barely resolved in figs. 1c, 1d). Note that due to the almost exact compensation of the interaction of e^- with D^0, for $m=0, 1, 2$ these asymptotical values are practically attained at $M\leqslant 3$–5. (It is interesting to note that the states of physically the same origin emerge with increasing M in the spectra of a 2D magnetoexciton bound to the ionized donor (D^+, X) –

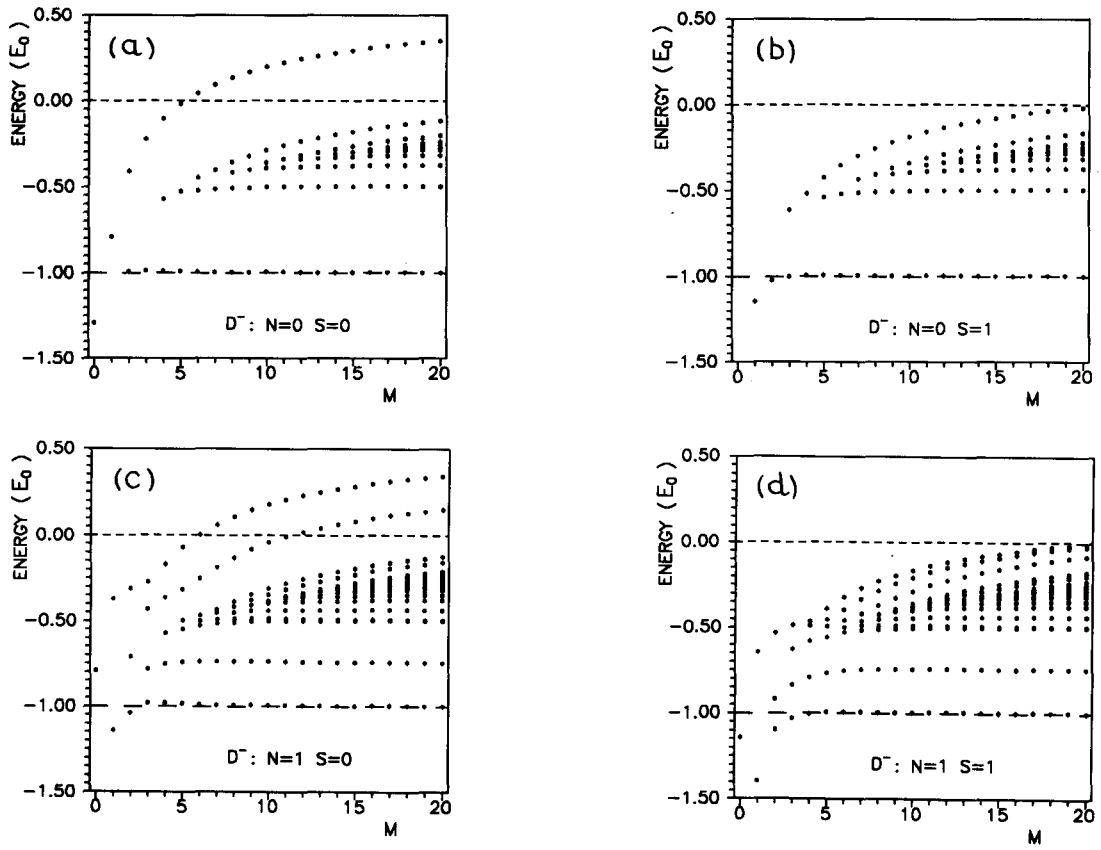

Fig. 1. Interaction energies (in units of $E_0=(\pi/2)^{1/2}e^2/\epsilon r_H$) of D^- states with quantum numbers $M=N-M_z\leqslant 20$: (a) of the singlet $S=0$ states in the zero Landau level $N=0$; (b) of the triplet $S=1$ states in the $N=0$ Landau level, the state with $M=0$ is absent due to the Pauli exclusion principle; (c) of the singlet $S=0$ states in the $N=1$ Landau level; (d) of the triplet $S=1$ states in the $N=1$ Landau level. The states of D^- lying below the ground state of D^0 with the energy $-E_0$ (shown by the dashed line) are stable against the removing of an electron.

when a hole leaves D^0, the latter being in the excited state $m=1, 2, \ldots$ [21]. The difference between D^- and (D^+, X) is that for a magnetoexciton such states lie in the continuum – the exciton band with the delocalized wave functions – and are the *resonant* states with finite widths.)

Now let us turn to the arising excited branches of the spectra. Physically, they emerge when the angular momentum of the center of mass is increased while the angular momentum of relative motion is fixed low so that two electrons in close proximity to each other are moved away from the donor ion. The energies of these branches are approximately given by $\mathscr{E}(S) - \text{const} \times M^{-1/2}$, where $\mathscr{E}(0) = 0.7071 E_0$ ($\mathscr{E}(1) = 0.3536 E_0$) are the upper bounds of the spectra of two interacting electrons in the singlet $S=0$ (triplet $S=1$) states with $N=0$.

Between these two limiting cases of the low-lying and the excited states, there are also intermediate cases corresponding to the near continuum spectrum, when two electrons are distributed over larger areas weakly interacting with each other and the donor ion.

(2) Let us now consider the stability of D^- states with respect to a dissociation $D^- \rightarrow D^0 + e^-$. In the $N=0$, 1 Landau levels an electron is most tightly bound to the donor ion in the state $n=0$, $m=0$, the corresponding binding energy of a 2D neutral donor D^0 is given by [23] $V_{0,0} = E_0$ (see also ref. [20], where in a 2D situation arbitrary fields H are considered). Hence, the states of D^- are stable when the corresponding interaction eigenenergy lies below $-E_0$. The states with energies exceeding $-E_0$ are the autoionization states unstable against the removing ion of an electron. Notice though that strictly speaking, there is no continuum and all states are localized.

For the singlet states of D^- with electrons in the zero Landau level $N=0$, there is only one stable state with $M=0$ and the binding energy $E_b = 0.2929 E_0$. Let us compare this result with the high-accuracy quantum Monte Carlo calculation [11] of the singlet s-ground state of D^- in the high field regime $\hbar\omega_c / 2\,\text{Ry}^* = 3$ ($H \simeq 20.2$ T). For GaAs a QW of width 100 Å [11] gives (i) 1.13 Ry* when the differences between the effective masses m_1^*, m_2^* and dielectric constants ϵ_1, ϵ_2 of GaAs and $Ga_{1-x}Al_xAs$ are not taken into account, (ii) 1.11 Ry* for $m_1^* \neq m_2^*$ and $\epsilon_1 \neq \epsilon_2$ and (iii) 1.22 Ry* for $m_1^* \neq m_2^*$, $\epsilon_1 \neq \epsilon_2$ and the non-parabolicity of the GaAs conduction band is taken into account. Our strictly 2D result $0.2929 E_0 \simeq 1.27$ Ry*, as one might expect, is higher.

Among the triplet $N=0$ states there are three stable states from which only one ($M=1$) is strongly bound with $E_b = 0.1465 E_0$. The singlet s-state (not taking account of spin energies in H) has the lowest *full* energy as it must be: the corresponding wave function has no nodes. The difference between the eigenenergies of the singlet and the triplet ground states $\Delta E_{ST} = 0.1465 E_0$. In a strong magnetic field the Zeeman spin splitting $|g| \mu_B H > \Delta E_{ST} \propto \sqrt{H}$ so that the spin-polarized triplet is the global ground state. Note also that the largest binding energy of the donor-bound 2D magnetoexciton (D^+, X) in zero Landau levels $0.1189 E_0$ [21] turns out to be lower then the binding energies of both the singlet and triplet D^- states.

In the case when one electron is excited to the $N=1$ Landau level there are two stable singlet states (the only strongly bound one is with $M=1$ and $E_b = 0.1416 E_0$). For the triplet $S=1$ there are five stable states, among them two states are strongly bound: (i) with $M=0$, $E_b = 0.1465 E_0$ (which strictly coincides with the binding energy of the triplet ground state $M=1$, $N=0$ – as it should be, since the two states are $t \rightarrow -t$ counterparts) and (ii) with $M=1$ and the largest binding energy $E_b = 0.3965 E_0$.

It is interesting to elucidate the possibility in a strong magnetic field H for the donor ion to bind *three* electrons forming stable D^{2-} states. As it turns out, both the D^{2-} doublet $S=\frac{1}{2}$ and the quadruplet $S=\frac{3}{2}$ are unstable against the dissociation via $D^{2-}_{S=1/2(3/2)} \rightarrow D^-_{S=0(1)} + e^-$. Details of the calculations will be presented elsewhere [22].

(3) Let us derive the energies of transitions with the absorption of an infrared photon which excites one electron from zero to the Nth Landau level. Both for the singlet and the triplet one should take account only of transitions from the most tightly bound $N=0$ ground states with $M=0$ and $M=1$, respectively (see fig. 2). Other stable triplet states have such low binding energies, that even at helium temperatures they are thermally ionized.

In the Faraday geometry dipole-allowed are the transitions conserving S, S_z and with $\Delta M_z = \pm 1$. When the admixture of higher Landau levels is neglected, from the $N=0$ D^- ground states only the

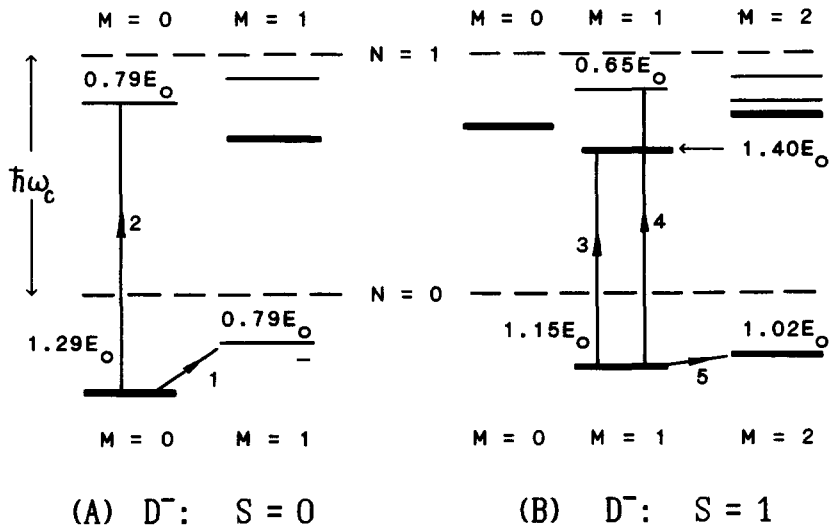

Fig. 2. Electric dipole-allowed transitions (a) from the singlet s-ground state of D^-, (b) from the triplet p-ground state of D^- ($M=N-M_z$). The energies of the free $N=0$, $N=1$ Landau levels are shown by dashed lines (disregarding the spin energies $g\mu_B HS_z$). Thick (thin) solid lines denote the stable (unstable) states of D^-. The energies of transitions are: $\hbar\omega_1=0.5E_0$, $\hbar\omega_2=\hbar\omega_4=\hbar\omega_c+0.5E_0$, $\hbar\omega_3=\hbar\omega_c-0.25E_0$, $\hbar\omega_5=0.13E_0$. Transitions (1), (5) with $\Delta M\neq 0$ are weak (see text). Mixing between states with $\Delta M_z=\pm 1$ due to the electric field of ionized donors is not taken into account. Other weak transitions and further possible transitions from the stable final states to higher Landau levels are not shown.

$\Delta M_z=1$ transitions with $\Delta N=1$ and conserving M are allowed. When the mixing is included, transitions with $\Delta M\neq 0$ ($\Delta N\neq 1$ and $\Delta N=1$, $\Delta M\neq 0$) become weakly allowed as $(E_0/\hbar\omega_c)^2$. Examples of such transitions are the $D^-\rightarrow N=0$ s$\rightarrow$p photoionization transition of the singlet with energy $\hbar\omega_1=0.5E_0$ (corrections $\sim E_0^2/\hbar\omega_c$ are neglected) and the p$\rightarrow$d transition of the triplet with $\hbar\omega_5=0.13E_0$ (see fig. 2).

Consider now the strong $\Delta M=0$ transitions. For $N=1$ there is only one singlet state with $M=0$ which is unstable. Hence, the transition to it leads to the photoionization of D^-; the photon energy $\hbar\omega_2=\hbar\omega_c+0.5E_0$ (see fig. 2a). For the triplet $S=1$ with $N=1$, there are *two* states with $M=1$. Since the two initial basis states from (4) are exactly degenerate (they are $t\rightarrow -t$ counterparts), the corresponding exact eigenfunctions are given by

$$\frac{\phi_{0,1}(\boldsymbol{r})\phi_{1,0}(\boldsymbol{R})\pm\phi_{1,0}(\boldsymbol{r})\phi_{0,1}(\boldsymbol{R})}{\sqrt{2}}=|z_1|^2-|z_2|^2, \tag{6a}$$

$$=z_1^* z_2-z_1 z_2^* \tag{6b}$$

(we omit for brevity the normalization factors and the exponential parts of the wave functions). Transitions to both states (6) are dipole-allowed and have *equal values* of the transition matrix element: dipole-allowed is the transition $\phi_{0,1}(\boldsymbol{r})\phi_{0,0}(\boldsymbol{R})\rightarrow\phi_{0,1}(\boldsymbol{r})\phi_{1,0}(\boldsymbol{R})$ with the conservation of the quantum numbers of the relative motion of the two electrons and with $\Delta n=1$, $\Delta m=0$ for the quantum numbers of the center-of-mass motion.

Consider first the transition to the state (6a) which is stable. The energy of the infrared photon is $\hbar\omega_3=\hbar\omega_c-0.25E_0$ (see fig. 2b), i.e. it falls below the cyclotron energy $\hbar\omega_c$. To the best of our knowledge, no experimental evidence for this transition is as yet available. Presumably this is due to saturation effects, so the time-resolved experiments or lower FIR intensities would be promising. It should be stressed that the state (6a) has a large binding energy $E_b=0.3965E_0$, hence this transition presumably cannot be detected by the magnetophotoconductivity measurements.

Now let us turn to the second state (6b) which is *unstable* (interaction energy $-0.6465E_0$). Therefore, transition to it leads to the photoionization of

D^-, and the corresponding energy of the photon is $\hbar\omega_4=\hbar\omega_c+0.5E_0$, which strictly coincides with the above considered case of the singlet state (this "accidental" degeneracy would be lifted with the admixture of higher levels).

In conclusion, for a strictly 2D case and in the strong magnetic field approximation, we have considered the singlet and triplet D^- spectra and the energies of the dipole-allowed transitions. In particular, we have shown that the energies of photoionization transitions exceed the binding energies of D^- which may explain the discrepancy between the calculations [11] and the experimental values [9] derived from the $D^-\rightarrow N=0$, 1 spectra. For a quantitative description of the experimental situation, mixing between Landau levels and the motion perpendicular to a QW are to be included into the present consideration. This work is now in progress.

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