Exact solutions for barrier D^- states at high magnetic fields

Herbert L. Fox and David M. Larsen
University of Massachusetts–Lowell, Lowell, Massachusetts 01854
(Received 3 November 1994)

Solutions which are exact in the high-magnetic-field limit have recently been reported for a two-dimensional D^- problem in which the two electrons and the fixed positive ion are all confined to the x-y plane. The magnetic field is applied in the z direction. We have generalized this model by allowing the fixed positive ion to reside on the z axis at a distance ζ from the x-y plane; the electrons remain confined in that plane. We call this configuration a barrier D^- center. Although exactly four bound D^- states exist at $\zeta=0$, we find that there are ten different states which, at fixed magnetic field, become bound in various intervals of ζ values; of these states as many as eight can coexist for the barrier D^- in certain ranges of ζ. Remarkable changes in the symmetry of the barrier D^- ground-state wave function are found, the progression of bound ground states being given by $M=0$ singlet $\rightarrow M=-1$ triplet $\rightarrow M=-2$ singlet $\rightarrow M=-3$ triplet $\rightarrow M=-5$ triplet as ζ increases from zero at fixed magnetic field. (M is the orbital z angular momentum quantum number.) At sufficiently large values of ζ all bound states of the barrier D^- center disappear. This progression leads to a predicted magnetic vaporization of barrier D^- centers.

I. INTRODUCTION

Exact solutions to quantum-mechanical problems are often of great interest even though they may pertain to model systems which cannot be very accurately realized in experiment. An example is the problem of a strictly two-dimensional D^- center in the presence of a strong perpendicular magnetic field. This center consists of a fixed positive ion and two electrons, all confined to the x-y plane. In the limit of an infinite magnetic field perpendicular to the plane, the D^- wave function can be constructed exclusively from free-electron $N=0$ Landau-level basis functions and exact eigenvalues and eigenvectors can be obtained.[1–3]

The D^- eigenstates can be classified according to the total orbital angular momentum of the electrons along the z direction, $\hbar M$, and the permutation symmetry of the spatial wave functions upon interchange of electron coordinates (singlet and triplet, corresponding to symmetric and antisymmetric space wave functions, respectively). D^- states are bound if the minimum energy required to remove one of the two electrons to infinity without flipping a spin is positive. In that sense, exactly four bound states are found for the two-dimensional D^- center in the infinite field limit. This set of bound states is comprised of an $M=0$ singlet ground state and three excited triplet states with $M=-1$, -2, and -3. In addition there exists a manifold of unbound but, nevertheless, localized excited states.

This two-dimensional model can give us qualitatively important information on the behavior of D^- centers located in the middle of narrow quantum wells. Suppose, however, that the positive ion, instead of being fixed at the center of the well, is located near the barrier edge or inside the barrier. Can bound D^- centers form in that case? If so, how strongly bound might such centers be?

To gain some insight into such questions, we have generalized the two-dimensional D^- model by retaining electron confinement in the x-y plane, but moving the positive ion out of the plane and fixing it a distance ζ along the z axis. The two-dimensional model is just the $\zeta=0$ case of our generalized model. We call the system which results when $\zeta>0$ a "barrier D^- center." We shall find asymptotically exact solutions for the barrier D^- problem as functions of ζ. Before discussing these solutions in detail, we offer some qualitative comments.

It is clear that as ζ increases from zero, the attractive interaction responsible for binding decreases. However, the binding energy of the D^- center depends on the difference between the energy of the D^- center and that of the neutral donor (D^0) left behind when one electron is removed from the D^- ion. Both the total (two-electron) binding of the D^- ion and that of the D^0 decrease as ζ increases. The pertinent question is which decreases faster. For this reason one cannot rule out, without examination, the possibility that the D^- center could have more bound states for some nonzero value of ζ than for $\zeta=0$.

It is to be expected that the qualitative nature of the lowest-lying D^- states will change with increasing values of ζ. Coulomb attraction terms in the D^- Hamiltonian decrease in strength as ζ increases, whereas the electron-electron repulsion term is independent of ζ. Thus, at larger values of ζ, the repulsion becomes relatively more important, and the electrons can lower their energy most effectively by forming highly correlated states in which they are well separated in position. (Correlation between electrons is relatively weak in the bound $\zeta=0$ eigenstates.) We shall show by explicit calculation that at the higher ζ values strong angular correlation exists between the electrons in the lower-lying states, correlation which tends to keep the electrons on opposite sides of the origin (the origin is taken in this paper, as the projection of the positive ion on the x-y plane). Strong correlation cannot occur in states with $M=0$ or -1. Thus, it is not surprising that D^- ground-state wave functions at the larger ζ

0163-1829/95/51(16)/10709(5)/$06.00

values are associated with values of $|M|$ greater than 1. In general, states with relatively large values of $|M|$ offer the possibility of achieving a relatively high degree of correlation and hence increased spatial separation between electrons.

From classical considerations, presumably relevant to our problem at sufficiently large values of ζ, it would seem that the achievement of even an extreme degree of correlation between electrons would not suffice to produce binding. If we consider the classical electrostatics problem associated with our model, wherein two electrons, taken as classical point charges in the x-y plane, are in electrostatic equilibrium with a positive ion of charge $|e|$, which is at a distance ζ on the z axis from the plane, we find that *negative* work is required to move one electron to infinity and the other to the origin. In this classical case, we have an extremely highly correlated electron system, both electrons being equidistant from the origin and lying on a straight line passing through it. Nevertheless the system is unbound in the sense of this paper. In fact, consistent with this discussion, we find no bound states in our quantum-mechanical problem when ζ exceeds a certain critical value.

In the next section, we describe our method of calculation in brief and present and discuss our results.

II. CALCULATIONS AND RESULTS

The procedure which we employ here for finding asymptotically exact eigenvectors and eigenvalues is very similar to that already described in Ref. 1. We introduce the barrier D^- Hamiltonian, H, in donor atomic units (energies and lengths in units of the bulk donor Rydberg, R, and Bohr radius, a, respectively),

$$H = H_D(1) + H_D(2) + 2/|\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2| \ , \tag{1}$$

where $H_D(j)$ is the barrier donor Hamiltonian for electron j ($j=1$ or 2) given by

$$H_0(j) = -\nabla_j^2 + \frac{\gamma}{i}\frac{\partial}{\partial \varphi_j} + \tfrac{1}{4}\gamma^2\rho_j^2 - \gamma \ , \tag{2}$$

$$H_D(j) = H_0(j) - 2/(\rho_j^2 + \zeta^2)^{1/2} \ , \tag{3}$$

where $\boldsymbol{\rho}_j$ is the displacement of the jth electron from the origin, φ_j is the angle between $\boldsymbol{\rho}_j$ and the x axis, and γ is the conventional dimensionless magnetic-field strength defined by

$$\gamma = \hbar\omega_c/2R, \quad \omega_c = eB/mc \ .$$

Here, B is the strength of the uniform applied magnetic field along z and m is the conduction-band mass. Because γ is subtracted on the right-hand side of Eq. (2) the lowest eigenvalue of the free-electron Hamiltonian, H_0, is 0. (This defines our zero of energy.) The degenerate set of free-particle wave function with this eigenvalue have wave functions of the form

$$X_M(\rho) = (\gamma^{1/2}\rho)^{|M|}\exp(iM\varphi)\exp(-\gamma\rho^2/4) \times \frac{\gamma^{1/2}}{[2\pi(2|M|)!!]^{1/2}} \ , \tag{4}$$

where M, the quantum number for the z-orbital angular momentum, is a negative integer or zero. All other eigenfunctions of H_0 have eigenvalues equal to $2N\gamma$, where N is an integer greater than zero. In the limit of infinite field the eigenfunctions of Eq. (2) given in Eq. (4) are also eigenfunctions of H_D, as described in Ref. 1. The eigenvalues of H_D are given by

$$E_D(|M|) = \langle X_M(\rho)|-2/(\rho^2+\zeta^2)^{1/2}|X_M(\rho)\rangle \ , \tag{5}$$

and depend essentially upon the quantity α defined by

$$\alpha = \gamma^{1/2}\zeta \ . \tag{6}$$

Recursion relations for evaluating $E_D(|M|)$ are given in Eq. (11).

In Fig. 1, we plot $E_D(|M|)$ vs α for $0 \le |M| \le 5$. These energies approach each other as α increases, converging to a common limiting value of $-2/\zeta$ in the limit $\alpha \to \infty$. This behavior is very important and is easy to understand from the geometry of the wave functions. An electron in the state X_M is basically localized in an annular region centered on the origin and with a mean annular radius, which increases with increasing $|M|$. For large values of α, there is only a relatively small change of Coulomb potential as one moves from the mean annular radius associated with state X_M to that associated with state X_{M+1}.

We are interested in solutions exact in the limit $\gamma \to \infty$ with α fixed. As in Ref. 1, we introduce D^- basis functions given by

$$\Phi^{\pm}(M_1, M_2) = X_{M_1}(\rho_1)X_{M_2}(\rho_2) \pm X_{M_1}(\rho_2)X_{M_2}(\rho_1) \ , \tag{7}$$

where the + sign is for singlet states and the − sign for triplet states. The wave function displayed in Eq. (7) is a

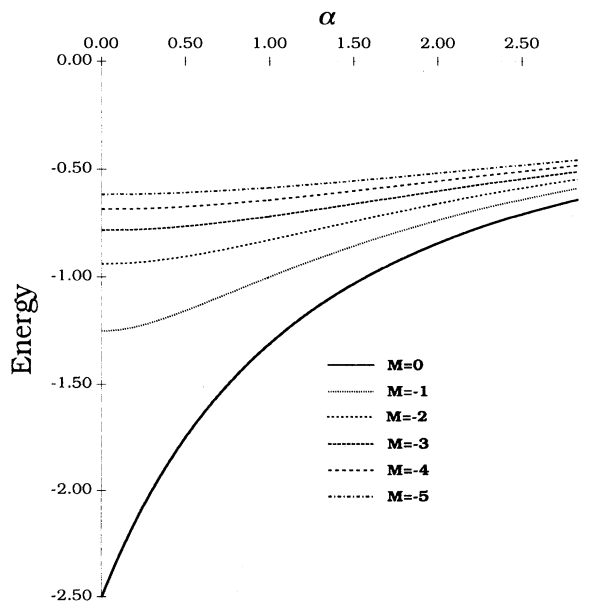

FIG. 1. Lowest energy, $E_D(M)$, of two-dimensional neutral donor orbitals for each value of M in the infinite field limit. The abscissa, α, is the distance of the positive ion from the x-y plane multiplied by $\gamma^{1/2}$. Energies are in units of $\gamma^{1/2}R$.

basis function with total orbital angular momentum M given by $M=M_1+M_2$. Since the total orbital angular momentum is a good quantum number of H, the most general form possible for an eigenfunction of H with total angular momentum M is

$$\Psi^{\pm}(M)=\sum_{M_1=0}^{M} C(M_1,M-M_1)\Phi^{\pm}(M_1,M-M_1)\ . \tag{8}$$

The corresponding eigenvalues, $E^{\pm}(M)$, satisfying

$$H\Psi^{\pm}(M)=E^{\pm}(M)\Psi^{\pm}(M) \tag{9}$$

can be found by solving the appropriate secular determinant. Binding energies associated with these eigenvalues were determined from

$$\varepsilon_B^{\pm}(M)=E^{\pm}(M)-E_D(0)\ . \tag{10}$$

Matrix elements required for evaluating $E^{\pm}(M)$ were calculated analytically. Those associated with the attractive potential, $E_D(M)$, given by Eq. (5), are found from the recursion relations,

$$\begin{aligned} &E_D(0)=-(2\pi\gamma)^{1/2}e^{\alpha^2/2}\mathrm{erfc}(\alpha/\sqrt{2})\ , \\ &E_D(1)=0.5(1-\alpha^2)E_D(0)-\gamma^{1/2}\alpha\ , \\ &E_D(|M|)=[(2|M|-1-\alpha^2)E_D(|M|-1)+\alpha^2E_D(|M|-2)]/(2|M|)\ \text{ for } |M|\geq 2\ . \end{aligned} \tag{11}$$

Off-diagonal elements of the secular determinants, matrix elements of the repulsive potential, were evaluated by writing the electron wave functions in relative and center-of-mass coordinates.[3] Binding energies corresponding to the lowest eigenvalue found for each secular determinant are plotted in Figs. 2 and 3.

We neglect throughout this paper Zeeman energies associated with the electron spins. The ground-state wave function at each value of α is the wave function associated with the most negative binding energy at that α value. Examination of Figs. 2 and 3 indicates that the sequence of ground-state wave functions for the barrier D^- center is $M=0$ singlet $\rightarrow M=-1$ triplet $\rightarrow M=-2$ singlet $\rightarrow M=-3$ triplet $\rightarrow M=-5$ triplet as α increases from zero. As many as eight bound states coexist for α between 0.41 and 0.48 and also between 0.80 and 1.16. No bound states are found for $\alpha>2.44$.

Figure 4 shows in polar plots the probability density of

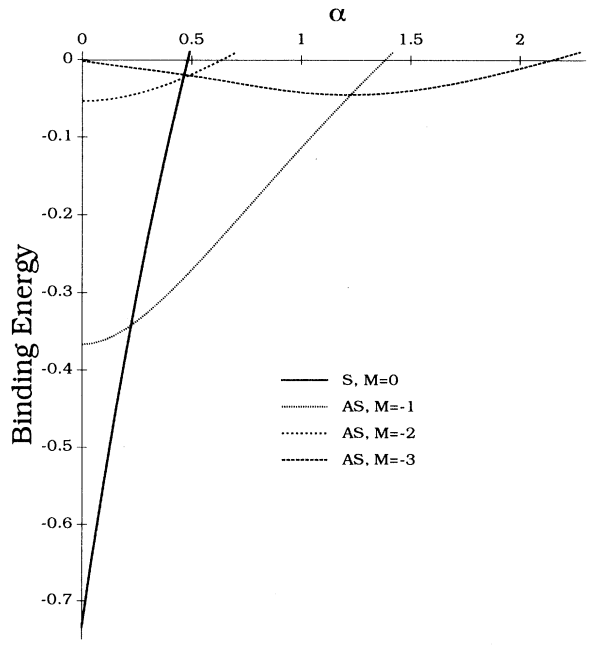

FIG. 2. Binding energies vs displacement, α ($=\gamma^{1/2}\xi$), of the positive ion from the x-y plane. Only those D^- states which do not become unbound as $\alpha\rightarrow 0$ are shown. The symmetric (singlet) state is labeled S, the antisymmetric (triplet) states AS. Binding energies are in units of $\gamma^{1/2}R$.

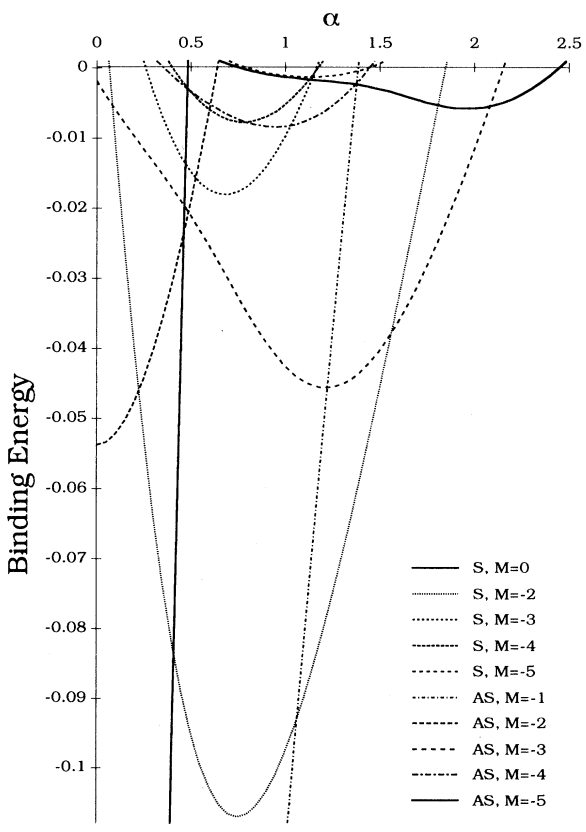

FIG. 3. Binding energies vs displacement, α ($=\gamma^{1/2}\xi$), of the positive ion from the x-y plane. All D^- states which bind for at least some values of α are shown. S and AS refer to symmetric (singlet) and antisymmetric (triplet) states, respectively. Notice the magnification of the energy scale relative to that of Fig. 2. Binding energies are in units of $\gamma^{1/2}R$.

finding the angle φ_{12} between ρ_1 and ρ_2 in typical singlet wave functions. Increases in angular correlation with increasing α are evident in a comparison of the dashed curves ($\alpha=0$) and the corresponding solid curves. These latter are characterized by a relatively large probability of finding φ_{12} in the second or third quadrant, indicating an increased preference at larger values of α for the electrons to reside on opposite sides of the projection of the positive ion on the x-y plane. The reason that strong angular correlation is achieved for the lowest-lying states when α is relatively large is that there is relatively little attractive-energy advantage for an electron to occupy the $M=0$ state as compared to a state of higher $|M|$ value, whereas, by occupying with significant probability the latter states, the electrons can reduce their mutual repulsion by establishing strong angular correlation favoring values of φ_{12} close to π. At low values of α, on the other hand, the $M=0$ state is so much lower in energy than any other level that the lowest-lying states all have at least one electron in the $M=0$ level, and angular correlation is inhibited.

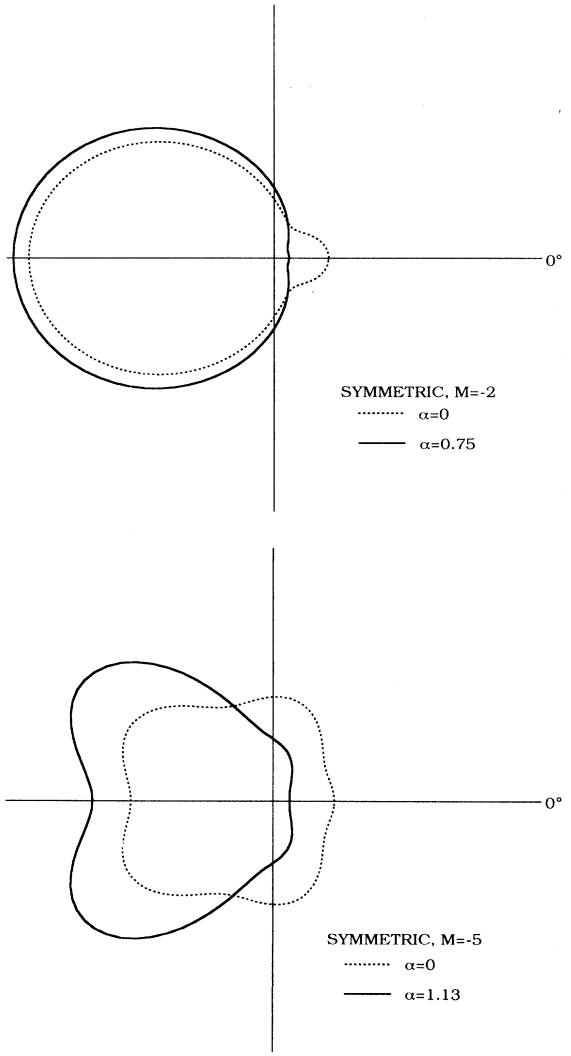

FIG. 4. Polar plots of the probability density of finding an angle φ_{12} between the displacement vectors of the two electrons in two typical D^- singlet states. The probability density represented by a point is proportional to the distance of that point from the origin. All graphs are plotted using the same scale to facilitate visual comparisons. The value of α chosen for each solid curve approximates the displacement of the positive ion at which maximum binding energy occurs for the state associated with that curve.

Magnetic-field-induced changes in the ground-state symmetry of D^- and related systems have been reported previously. Such effects have been predicted from variational calculations in a positronium atom bound to a proton,[4] and in D^- ions located near to or inside the barriers of quantum wells.[5] Closer in spirit to the present work, however, is the recently reported exact numerical solution of the problem of two electrons in the x-y plane bound in a harmonic-oscillator potential in the presence of a magnetic field in the z direction (a model proposed for the quantum dot problem).[6,7]

In the quantum dot model,[6,7] an infinite sequence of ground states for a fixed dot size but increasing magnetic field is found. Starting at zero field with an $M=0$ ground state, the M values attached to the ground-state wave function decrease without limit in single-unit increments as the magnetic field increases to infinity. Even values of M are associated with singlet states, odd values with triplet states. Increasing the magnetic field leads to more and more highly correlated ground-state electronic wave functions, the electrons tending to lie diametrically opposed on opposite sides of the center of the quantum dot and to press up against the harmonic-oscillator potential barrier, keeping as far away from each other as possible without climbing too high on the barrier. The increase of ground-state $|M|$ values with field at high fields is simply a consequence of the shrinkage of the cyclotron radius with increasing field, an effect which would draw the electrons closer to the center (and hence to each other) if not counteracted by an increase in $|M|$. All states are bound since infinite energy is required to remove a quantum-dot electron to infinity.

Experimental conditions favoring formation of barrier D^- ions of the type discussed here are not hard to imagine. Consider a GaAs quantum-well structure with very wide barriers and undoped narrow wells in which the barriers are selectively Si doped both in the center and near the barrier-well interface. The center doping would provide electrons to the well, which, in the absence of doping near the barrier-well interface, would be unbound (at large distances from the well, the center doping should act like a uniform sheet of positive charge). These unbound electrons could be trapped by donor ions near the interface to form barrier D^- centers. At "high" magnetic fields, the results of this paper should give a good qualitative description of the electronic structure of the states present.

A remarkable feature of the barrier D^- ions is that under certain conditions, one can expect magnetic "vaporization" of these centers. At $\zeta=0$ or in bulk semiconductors, increasing magnetic fields deepen the ground state of shallow impurities thereby promoting magnetic localization or "freeze out" of electrons onto impurity sites. In

contrast, for barrier D^- states increasing the magnetic field at appropriate fixed values of ζ leads to increasing values of α [see Eq. (6)] and eventual loss of D^- binding. In principle, such behavior could be detected by low-temperature magnetotransport measurements in quantum-well systems of the type described above.

Regarding variational calculations of barrier D^- centers in real quantum wells, the present results suggest that for the highly correlated states, namely, the $M=-2$ symmetric state and states with $|M|>2$, Chandrasekar-type wave functions, which are often used in D^- calculations, may give relatively poor energy estimates.

[1]D. M. Larsen and S. Y. McCann, Phys. Rev. B **45**, 3485 (1992).
[2]A. B. Dzyubenko, Phys. Lett. A **165**, 357 (1992).
[3]A. H. MacDonald, Solid State Commun. **84**, 109 (1992).
[4]D. M. Larsen, Phys. Rev. B **20**, 5217 (1979).
[5]S. Y. McCann, Ph.D. thesis, University of Massachusetts, 1993.
[6]U. Merkt, J. Huser, and M. Wagner, Phys. Rev. B **43**, 7320 (1991).
[7]M. Wagner, U. Merkt, and A. V. Chaplik, Phys. Rev. B **45**, 1951 (1992).