Negative hydrogenic ion or D^- center in a quantum wire

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(Received 24 June 2005; published 3 October 2005)

The negative hydrogenic ion is not only of physical but also historic importance for it is the first quantum mechanical three-body problem that involves the intrinsic electron-electron correlation. Recent advances in nanotechnology have brought the study of this system in a quantum wire into the forefront. The nonanalytic behaviors of the binding energy $\epsilon_b^{2\text{nd elec}}$ of the second electron in a negative hydrogenic ion in one dimension and in a quantum wire (QWR) as a function of the Coulombic repulsive strength γ and the wire radius b are first identified. They are then exploited, together with the recognition of the well-known near-infinite (à la Loudon) binding energy of the neutral hydrogenic atom, to set up a model that calculates $\epsilon_b^{2\text{nd elec}}$ in a QWR directly, thereby avoiding the pitfalls of a variational approach to this problem. This $\epsilon_b^{2\text{nd elec}}$, found to be 0.4 Ryd, is an exact solution for a wire whose radius is nearly zero. For a finite b, it is shown that still $\epsilon_b^{2\text{nd elec}} \approx 0.4$ Ryd, which is independent of b as long as $0 < b \ll a_{\text{Bohr}}$ due to the inverse square nature of the Coulomb force. Its comparisons with the corresponding cases in three dimensions and two dimensions is discussed.

DOI: 10.1103/PhysRevB.72.153303 PACS number(s): 73.21.Hb, 71.55.−i, 73.90.+f

The advance in nanotechnology has not only resulted in many exotic applications[1] but also stimulated many theoretical and experimental studies of quantum confinement, including that of the binding energy of a donor (a D^0 center) in a quantum well,[2–5] and of a D^- ion in two-dimensional semiconductors.[6–11] Recent progress in lithographic and growth techniques have also made possible the fabrication of quantum wires (QWRs) with radii less than 100 Å (Refs. 12–14), resulting in applications such as quantum wire lasers.[1] In contrast to the numerous theoretical works on the neutral D^0 states in quasi-one-dimensional systems (i.e., QWRs) based on the variational approach,[15] and on the two-scale effective potential approach,[16] there has been relatively little work on the D^- ion in a QWR.[17] We shall start our investigation by explaining the reason why this is so.

In the pioneering work of Loudon on a one-dimensional hydrogen atom[18] the Schrödinger equation takes the form $d^2\phi/dz^2+\alpha\phi/|z|-\phi/4=0$ in the variable $z \equiv x/(\alpha a_0/2)$, which is defined in terms of the unknown length scale αa_0, with the energy eigenvalue denoted now by $\epsilon \equiv -\hbar^2/[2m^*(a_0\alpha)^2]$. Here m^* is the effective electron mass and $a_0=\hbar^2\kappa/m^*e^2$ is the usual effective Bohr radius,[5] κ being the dielectric constant. It is then immediately obvious that setting $\alpha=0$ would yield the ground-state energy, and its wave function is given by $\phi_0(x)=N\lim_{\alpha\to 0}\exp(-|z|/2)$, so that $[\phi_0(x)]^2=\delta(x)$, implying that the electron collapses onto the positive ion core. Actually the above is justified by a more detailed mathematical analysis with the use of a truncated Coulomb potential $V=-e^2/(|x|+a)$, provided we let $a\to 0$ at the end of the calculation. The relation between α and a is $\alpha=1/[2\ln(a_0/a)]$. This results in an infinitely negative ground-state energy

$$\epsilon_0 = \lim_{a\to 0}\frac{-2\hbar^2}{m^* a_0^2}\left[\ln\left(\frac{a_0}{a}\right)\right]^2 = \lim_{\alpha\to 0}\frac{-\hbar^2}{2m^*(a_0\alpha)^2} \to -\infty \quad (1)$$

or an infinite binding energy. Incidentally, one might wonder why there is no such solution of $\alpha=0$ for the three-dimensional (3D) hydrogen atom in spite of the fact that the Schrödinger equation for the radial wave function $\chi(r) \equiv rR^{3D}(r)$ is exactly the same as that for the one-dimensional (1D) function $\phi(x)$. The answer is somewhat subtle[18] and partially explained in Baym's book.[19] However, all the other solutions associated with the excited states of the 1D problem agree, one-to-one, with those associated with all the familiar S-states of the 3D radial problem, including the energy spectrum.

Let us also briefly review the spirit of the two-scale solution[16] of Jan, Lee, and Chuu for the problem of D^0 in a QWR of radius b, with $0 < b \ll a_0$. We can qualitatively divide the energy of the single electron attracted to the nuclear core into two parts, that of the longitudinal x motion along the wire and that of the radial ρ-motion transverse to the wire, $\epsilon \approx \epsilon_x + \epsilon_\rho$ where $\epsilon_\rho \sim T_\rho \sim \hbar^2/2m^*\rho^2$ and

$$\epsilon_x \approx (-2\hbar^2/m^* a_0^2)[\ln(a_0/b)]^2 \quad (2)$$

as given by Eq. (1) in which b has replaced the cutoff length a. This is because the Coulomb potential energy in our QWR with $\rho_{\max}=b$ is $V\sim -e^2/(|x|^2+b^2)^{1/2}$, which is qualitatively similar to the truncated potential in 1D,[18] $V=-e^2/(|x|+a)$ as $|x|\to 0$, with the radius b playing the role of a. Correspondingly,[16] the longitudinal spread of the wave function $\phi_0(x)$ is

$$\Delta x_0 \sim a_0/[2\ln(a_0/b)], \quad (3)$$

where $a_0/b \gg 1$.

We observe that as b approaches zero, ϵ_ρ diverges quadratically while ϵ_x diverges only logarithmically. It is therefore justified to factorize the ground-state wave function into the ρ-part and the x-part as in the Born-Oppenheimer adiabatic approximation. As we let b deviate from zero the dominating effect on the collapsing electron is its transverse expansion to fill up the cross section of the wire so that its ρ wave function is proportional to the Bessel function $J_0(k_0\rho)$,

with $J_0(k_0 b)=0$, while the accompanying spread in the longitudinal x direction is relatively minor.[16] To the leading order of b/a_0 in the wave function the latter spread can be ignored, i.e., the electron spreads out radially into a disk of radius b while remaining longitudinally in $\phi_0^2(x)\sim\delta(x)$ in a QWR.

We now turn to the D^- problem in exactly one dimension ($b=0$). The most important difference from the corresponding D^0 problem lies, of course, in the Coulomb repulsion term $V_c=\gamma e^2/r_{12}$ between the two electrons in the negative D^- ion. Here the coefficient γ is an artificial parameter to distinguish the interelectron repulsion from the electron-core attraction $-e^2/r_i$. The key point is that physically the expectation value $\langle V_c\rangle=\int_{-\infty}^{+\infty}|\Psi(\mathbf{r}_1,\mathbf{r}_2)|^2 V_c d\mathbf{r}_1 d\mathbf{r}_2$ must not be allowed to diverge to $+\infty$ for any two-electron wave function $\Psi(\mathbf{r}_1,\mathbf{r}_2)$. In the particular case of one dimension, since the integral around the origin $\int_{-\delta}^{+\delta}V_c(x_{12})dx_{12}=\int_{-\delta}^{+\delta}\gamma e^2/|x_{12}|dx_{12}\to\infty$ we must impose the boundary condition

$$\Psi(x_1,x_2)=0 \quad \text{at } x\equiv x_1-x_2=0 \tag{4}$$

as long as $\gamma>0$. This is no longer required in two or three dimensions for the corresponding integral $\int_{-\delta}^{+\delta}V_c(x_{12})x_{12}dx_{12}$ or $\int_{-\delta}^{+\delta}V_c(x_{12})x_{12}^2dx_{12}$ converges. We emphasize that Eq. (4) must be obeyed even at the cost of sacrificing a possible infinite attraction $-Ze^2/x_1\to-\infty$ or $-Ze^2/x_2\to-\infty$ as $|x_i|\to 0$. Although the attraction between the positive nuclear (or ionic) core and either of the two electrons maximizes as the electron approaches the core, physically the core radius $|x_i|_{\min}\gtrsim 10^{-13}$ cm, rendering the maximum attraction large but finite. On the other hand, the distance x between two pointlike electrons could approach zero without restriction, rendering the Coulomb repulsion genuinely divergent. In other words, we may allow an electron to touch the core, but not to touch the other electron. It follows that if one electron has collapsed onto the minute core, the other electron should not be allowed to do the same so as to avoid touching each other. (Mathematically, we are assuming $\Delta x_0\gtrsim$ core size.) Consequently the ground state of the D^- center is not an analytic function of γ at $\gamma=0$. When $\gamma=0$, its ground state is just a simple product $\Psi_{\rm grd}(x_1,x_2;\gamma=0)=\phi_0(x_1)\phi_0(x_2)$, with $E_{\rm grd}=2\epsilon_0$. Yet, when $\gamma=0^+$ is infinitesimally small but not zero, we have $\Psi_{\rm grd}(x_1,x_2;\gamma=0^+)=(1/\sqrt{2})[\phi_0(x_1)\phi_1(x_2)-\phi_0(x_2)\phi_1(x_1)]$, with $E_{\rm grd}=\epsilon_0+\epsilon_1$. This antisymmetric spatial wave function satisfies the boundary condition in Eq. (4) while remaining as an exact eigenfunction of the lowest energy of the unperturbed (i.e., $\gamma=0$) Hamiltonian. It must also be accompanied by a spin triplet function. The binding energy of the second electron is immediately identified as $\epsilon_b^{\rm 2nd\ elec}=-\epsilon_1=|\epsilon_1|=1$ Ryd$=\hbar^2/2m^*a_0^2$. We also recognize two vastly disparate length scales, namely, the practically zero length scale αa_0 for ϕ_0 and the familiar Bohr radius a_0 for ϕ_1, the latter being also roughly the separation between the two electrons.

If we follow the variational approach pioneered by Chandrasekhar[20] and widely used for calculating $\epsilon_b^{\rm 2nd\ elec}$ in quasi-two-dimensional (Q2D) quantum wells[6–9] by writing a trial ground-state wave function of the type

$$\Psi^{\rm trial}(x_1,x_2)=\phi_0(x_1)\phi_0(x_2)(1+C|x_1-x_2|+\cdots)$$

we might expect the variational parameter C to increase monotonically with γ so that a strong repulsion would result in a large separation between the two electrons. This same parameter C would also be expected to decrease continuously to zero as γ approaches zero so as to recover the correct limiting case of noninteracting electrons. The binding energy $\epsilon_b^{\rm 2nd\ elec}$ is then given by the difference $\epsilon_b^{\rm 2nd\ elec}=E_{\rm grd}(C\neq 0)-E_{\rm grd}(C=0)$. This approach would fail for QWRs for two reasons. First, as $\gamma\to 0^+$ we would never retrieve the noninteracting case due to the nonanalyticity discussed above. Second, since the two numbers $E_{\rm grd}(C\neq 0)$ and $E_{\rm grd}(C=0)$ are consequently unconnected, to calculate $\epsilon_b^{\rm 2nd\ elec}$ we must compute each of them separately. However, since Loudon's $E_{\rm grd}(C=0)=-\infty$,[18] a finite $\epsilon_b^{\rm 2nd\ elec}$ would have to result from the difference between two independent, infinitely large numbers.

Let us conclude our study of the D^- problem in 1D but now with $\gamma=1$. The two-electron ground-state wave function that obeys Eq. (4) must be of the form

$$\Psi_{\rm grd}(x_1,x_2)=\frac{1}{\sqrt{2}}[\phi_0(x_1)\psi(x_2)-\phi_0(x_2)\psi(x_1)] \tag{5}$$

in which ϕ_0 is there to lower the energy as much as possible while $\psi(x)=\Sigma_n a_n\phi_n(x)$ is a general single-particle orbital. We may exclude the term of $n=0$ from the above sum due to the antisymmetry in $\Psi_{\rm grd}(x_1,x_2)$ of Eq. (5). Consequently the scalar product $\langle\phi_0|\psi\rangle=0$. Thus one of the two electrons occupies the ϕ_0 orbital, with the other occupying the ψ orbital that is orthogonal to ϕ_0. Without further calculation we conclude immediately that $\epsilon_b^{\rm 2nd\ elec}=0$ now that the Coulomb repulsion is in full force, for any nuclear core (of charge $+e$) attraction felt by the second electron is now exactly cancelled by the full repulsion of the same magnitude exerted by the first electron sitting on that core, i.e., the nuclear core is effectively neutralized. This means the single-particle orbital $\psi(x)$ in Eq. (5) is just a plane wave representing a free particle at rest.

It is of crucial importance to point out that the electron wave packet of ϕ_0 and that of ψ are not only orthogonal to each other, but actually have no spatial overlap, because $\phi_0(x)\sim[\delta(x)]^{1/2}$ while $\psi(x)\sim xR^{\rm 3D}(x)$ so that $\phi_0(x)\psi(x)=0$.[18] In particular, the exchange-term contribution

$$E_{\rm exch}=\frac{e^2}{2}\int_{-\infty}^{+\infty}dx_1\int_{-\infty}^{+\infty}dx_2 \times\left(-2\phi_0(x_1)\psi(x_1)\phi_0(x_2)\psi(x_2)\frac{1}{|x_1-x_2|}\right)$$

to the total Coulomb energy $E_C=\langle\Psi_{\rm grd}|e^2/(|x_1-x_2|)|\Psi_{\rm grd}\rangle$ clearly vanishes as a result of the vanishing spatial overlap. So, only the direct or classical-like interaction between the two electrons is effective. The two electrons can now be treated as distinguishable like classical particles.

Finally we are in a position to tackle our main problem of a negative hydrogenic ion or a D^- center in a QWR of radius b, with $a_0\gg b>0$.

From what we learned previously about the D^- center when $b=0$, one of the two electrons will collapse onto the nuclear core in the state $\phi_0(x)$ while the other electron is left wandering in a single-particle orbital $\psi(x)$ that is not only orthogonal to but also has no spatial overlap with $\phi_0(x)$. As we let b deviate from zero, our experience in the D^0 problem[16] teaches us that predominantly each electron wave function would first expand radially to fill up the circular cross section so as to relieve the huge transverse kinetic energy otherwise pent up on the axis of the wire, while the modification of the longitudinal wave function is less pronounced. If the repulsion parameter $\gamma=0^+$, one electron e_1 would still remain in $\phi_0(x,\rho)\approx N_0 J_0(k_0\rho)\phi_0(x)$ $\approx N_0 J_0(k_0\rho)[\delta(x)]^{1/2}$ like an infinitely thin disk located at the origin, and the other electron e_2 would occupy $\phi_1(x,\rho)$ $\approx N_1 J_0(k_0\rho)\phi_1(x)$. For $\gamma=1$, we just have to replace $\phi_1(x,\rho)$ by $\psi(x,\rho)\approx N J_0(k_0\rho)\psi(x)$, in which $\psi(x)$ is yet to be determined. The spatial overlap

$$\int_{-\infty}^{+\infty}\psi(x)\varphi_0(x)F(x)dx \approx F(0)(\Delta x_0/a_0)^{1/2}\to 0 \qquad (6)$$

for any $F(x)$, where the width of the wave function $\phi_0(x)$ is $\Delta x_0\sim 0$ and that of $\psi(x)$ is of order a_0, rendering the two electrons still basically distinguishable from each other despite their transverse expansions.

Our job now is to determine from the Schrödinger equation the longitudinal wave function $\psi(x)$ for e_2, a negative circular charge of radius b with a radial distribution $J_0^2(k_0\rho)$, which is repelled by the negative e_1-charged-disk (of width Δx_0) as well as attracted by the nuclear point charge $+e$ at the center of this e_1-disk located at $x=0$. What must be ascertained first is whether the net potential $V_{12}(x)$ on the circular charge e_2 is negative or positive. Thus we need to calculate the work done by E_1 (produced by e_1 and the nuclear charge) in bringing the circular charge e_2 from ∞ to a finite separation $|x|$ from e_1 or, equivalently, the work done by the electric field E_2 (produced by e_2 alone) in bringing all the charges in the e_1-disk together with the positive nuclear center from ∞ to a finite separation $|x|$ from e_2. One convenient way to do the latter is to first bring every bit of e_1 initially distributed like $J_0^2(k_0\rho)$ over the disk to its positive nuclear center, thereby neutralizing the whole disk, then bring this neutralized disk from ∞ to $|x|$, and finally spread out the bits of e_1 radially away from the center to regain the initial distribution. The net work done by the E_2 field is only in the final step. This work is obviously positive, considering the qualitative pattern of the E_2-field lines produced by such a circular charge e_2, especially regarding their radially inward components. So, we conclude $V_{12}(x)$ is *negative* for any distance x between e_1 and e_2. This is significant for it means that the ground state $\psi_0(x)$ for e_2 must be a discrete bound state in the one-dimensional $V_{12}(x)$ potential well. It also marks a breakdown in analyticity in that the second electron was shown to be free when $b=0$ while it is now found to be bound with a finite binding energy for any $b>0$.

To evaluate $V_{12}(x)$, since the radius b is the only length parameter we can use the variable $x'=x/b$ to write $V_{12}(x)$

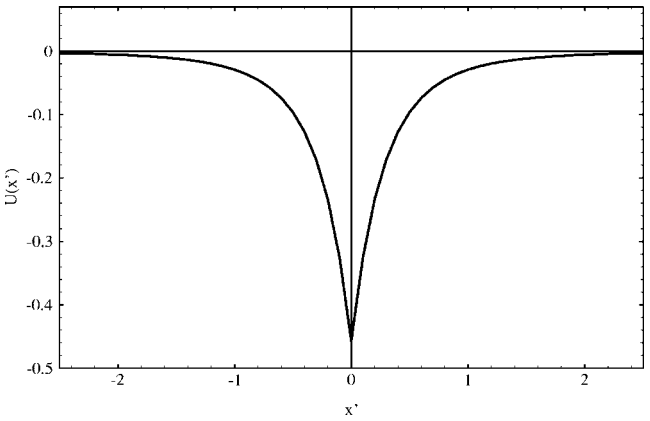

FIG. 1. A plot of the normalized potential $U(x')$ between the second electron and the nucleated charged disk of the first electron versus their normalized separation x'.

$=(2e^2/b)U(x')$ so that $U(x')$ is dimensionless and independent of b. Explicitly, writing the surface charge density as $\sigma(\rho)=(-e/\pi b^2)\sigma'(\rho)$, we have

$$U(x')=U_{\rm nucl}(x')+U_{\rm Coul}(x'), \qquad (7)$$

where

$$U_{\rm nucl}(x')=-\int_0^1 \sigma_2'(\rho_2^2+x'^2)^{-1/2}\rho_2 d\rho_2 \qquad (8)$$

and

$$U_{\rm Coul}(x')=\int_0^1 \rho_1 d\rho_1\int_0^1 \rho_2 d\rho_2 I(\rho_1,\rho_2;x') \qquad (9)$$

and

$$I(\rho_1,\rho_2;x')\equiv\frac{1}{\pi}\int_0^{2\pi} d\theta_1\sigma_1'\sigma_2'(x'^2+\rho_1^2+\rho_2^2 - 2\rho_1\rho_2\cos\theta_1)^{-1/2}. \qquad (10)$$

With the transverse wave function $J_0(k_0\rho)$ our dimensionless surface charge density is given by

$$\sigma'(\rho)=\pi J_0^2(k_0\rho)\Big/\left(2\pi\int_0^1 J_0^2(k_0\rho)\rho d\rho\right), \qquad (11)$$

where k_0 is such that $J_0(k_0 1)=0$ is the first zero of the Bessel function J_0. A plot of the numerically computed $U(x')$ versus x' is given in Fig. 1. If we denote the integrated area by $A\equiv\int_{-\infty}^{+\infty}U(x')dx'$ we can also obtain numerically $A=-0.318$.

A remark about the qualitative difference in $V_{12}(x)$ between the case of $b=0$ and that of $b=0^+$ is in order. We already know that when $b=0$, $V_{12}(x)=0$ even at $x=0$, since the charge $e_1=-e$ neutralizes exactly the $+e$ of the nuclear center, hence exerting no potential on e_2. On the other hand, let us consider $\lim_{x\to 0}V_{12}(x)$ for a small but finite wire, with the limit of $b\to 0^+$ taken only at the end of the calculation. In the limit of $|x|\ll b$, with the qualitative pattern of the electric field $E_2(x,\rho)$ produced by the circular charge e_2 taken into consideration, it is easy to show that $V_{12}(x)\sim -e^2/b<0$. This

is just as expected dimensionally, for b is the only length scale available now that $|x|\to 0$. Thus as $b\to 0^+$, $\lim_{b\to 0+}[\lim_{x\to 0} V_{12}(x)]\to -\infty$. On the other hand, since $U(x')\sim -(1/8|x'|^3)\to 0$ as $|x'|=|x|/b\to\infty$, it means $V_{12}(x)=(2e^2/b)U(x')\sim -(2e^2b^2/|x|^3)\to 0$ for any finite $|x|$ as $b\to 0^+$. In particular, $\lim_{x\to 0+}[\lim_{b\to 0} V_{12}(x)]\to 0$, indicating that the order of the two limits cannot be interchanged. Equivalently, $V_{12}(x)=2e^2A\,\delta(x)$ for $b\to 0^+$.

With $A<0$ it is well known that such an attractive δ-function potential well admits one and only one bound state whose binding energy is $\epsilon_b^{2\text{nd elec}}=m^*(2e^2A)^2/2\hbar^2$. More generally, for any weak one-dimensional potential well it is well known[21] that $\epsilon_{\text{grd}}=-(m/2\hbar^2)[\int_{-\infty}^{+\infty}V(x)dx]^2$. In our present case of the QWR this general formula yields exactly the same result as above. Adopting $A=-0.318$ we find

$$\epsilon_b^{2\text{nd elec}}=\frac{m^*(2e^2A)^2}{2\hbar^2}=4A^2\left(\frac{e^2}{2a_0}\right)=4A^2\ \text{Ryd}\approx 0.40\ \text{Ryd}. \tag{12}$$

That it is independent of the radius b as long as $0<b\ll a_0$ can be attributed to the inverse square nature of the Coulomb force law that renders $U(x')$ independent of b. The above result of $R\equiv\epsilon_b^{2\text{nd elec}}/\epsilon_b^{1\text{st elec}}=0.4\ \text{Ryd}/\infty\to 0$ is to be compared with $R=0.055\ \text{Ryd}/1\ \text{Ryd}\approx 5.5\%$ for 3D, and $R=0.48\ \text{Ryd}/4\ \text{Ryd}\approx 12\%$ for 2D,[6] and also with $R=0/\infty=0$ for $b=0$. The difference of infinite fold between 1D, Q1D, and 2D, 3D is striking. The physical reason is in two steps. First, the boundary condition of Eq. (2) leads strictly to $\epsilon_b^{2\text{nd elec}}=0$ for $b=0$. Then, as b deviates ever so slightly from zero, both electronic wave functions would predominantly expand to fill up the cross section of the QWR so as to relieve the pent-up transverse kinetic energy while the modification to the infinitely small longitudinal spread Δx_0 of the first electron that originally has succumbed to the strangle-hold of the positive nuclear core is minimal. This transverse expansion, however, relaxes the hermetic screening by the first electron of the central nuclear charge, leaving room for some net attraction exerted on the roaming second electron. Since the latter must be in a $\psi(x)$ orthogonal to $\phi_0(x)$ of the former, the separation between the two electrons must then necessarily be of order a_0, the only length scale other than Δx_0. This gives rise to $\epsilon_b^{2\text{nd elec}}\lesssim\text{Ryd}$.

Note that since the present $\psi_0(x)$ for the second electron depends only on $|x|$, it means $\Psi_{\text{Grd}}(1,2)=\Psi_{\text{Grd}}(2,1)$ is a spatially symmetric state, which must be accompanied by a spin singlet. It also implies that, in contrast to the case of $b=0$, $\Psi_{\text{Grd}}(x_1,\rho_1;x_2,\rho_2)\neq 0$ when $x_1=x_2$, because generally $\rho_1\neq\rho_2$ when $b>0$. Furthermore, since it is found that the condition $[(2m^*)^{1/2}/\hbar]\int_{-\infty}^{+\infty}[-V_{12}(x)]^{1/2}dx>\pi$ is not met, our $V(x)$ is not strong enough to bind another state.[22] This is consistent with the theorem of Hill.[23]

In summary, we have treated the negative hydrogenic ion in 1D ($b=0$) and Q1D ($0<b\ll a_0$) by setting up a model of two negatively charged ($-e$) disks, each of radius b, one always positioned at the positive nuclear core, the other roaming under the influence of the first disk together with its positive core. The binding energy of the roaming one is calculated directly and is found to be $\epsilon_b^{2\text{nd elec}}\approx 0.4\text{Ryd}$ for any b, as long as $0<b\ll a_0$. The ground state of the two-electron system is spatially symmetric, accompanied by a singlet spin state. A comparison with the corresponding cases in 2D and 3D is discussed. Generally speaking, when there are electrons and positive ion cores in a QWR, one electron will tend to collapse onto each core and hence reduce its charge by $1e$, rendering the molecular physics or chemistry qualitatively different from that in 2D and 3D.

The authors gratefully acknowledge the help of A. Sablauer for checking the numerical results.

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