

Transition from an artificial hydrogen molecule to a negative hydrogen ion D^-

Transición de una molécula artificial a un ión de hidrógeno D^-

Fulla Marlon¹ y Marín Jairo¹

Resumen. Se analizó el espectro energético de una molécula artificial de hidrógeno conformada por dos electrones, los cuales son liberados por dos impurezas donadoras al interior de un anillo cuántico bidimensional de espesor muy pequeño. Las dos donadoras están localizadas a lo largo del eje de simetría en dos posiciones diferentes respecto del centro del anillo. El complejo molecular se encuentra en presencia de un campo magnético uniforme orientado a lo largo del eje de las donadoras. Empleando el método de aproximación adiabática, se puede reducir el problema inicial de dos dimensiones a una ecuación de onda unidimensional con condiciones de frontera periódicas, la cual describe el movimiento rotacional de los electrones alrededor del eje de simetría. Se calculan algunos de los estados más profundos en función del radio del anillo, la intensidad de campo magnético y la separación entre donadoras. Mediante la variación de la posición de una de las impurezas, manteniendo la otra fija, es posible analizar la notable evolución del espectro energético desde una molécula hidrogenoide a un ión negativo hidrogenoide del tipo D^- .

Palabras clave: Nano-estructuras semiconductoras, Impurezas donadoras, Anillos cuánticos, Correlación electrónica, Molécula artificial.

Abstract. An artificial hydrogen molecule consisting of two electrons released by two on-axis donors into a two-dimensional quantum ring with a very small width is analyzed. The two donors are located at different positions respect to symmetry center. The molecular complex is in presence of an uniform magnetic field which is oriented along the donor axis. Following the well known adiabatic approximation, we can

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reduce the initial two-dimensional problem to a one-dimensional wave equation with periodic boundary conditions which describes the electron rotation around the symmetry axis. The some low-lying states are calculated as a function of the ring radius, the magnetic field strength and the donor-donor separation. By changing the position of one donor while the other one remains fixed is possible to analyze the remarkable evolution from a hydrogen-like molecule to a hydrogen-like ion energy spectrum.

Keywords: Semiconductors nanostructures, Shallow donor, Quantum rings, Electron correlation, Artificial molecule.

1. INTRODUCTION

The self-assembled quantum dots (QDs) obtained in the last few years (Lorke *et al.*, 2000), have been ideal solid-state structures to perform studies related to mesoscopic physics. They have long been thought of as artificial atoms or molecules since they are capable of confining a very small number of charge carriers. In this sense, these nanostructures do not only provide an ample opportunity of fabricating very promising electronic devices for high-tech applications, but also they can be used to dissect new atomic and molecular physical phenomena. In particular, the strong confinement of carriers in these structures facilitates the measurement of bound states energies up to room temperature of excitons, bi-excitons, and D⁻ ions, etc. That it is possible due to an increase of the bound states energies of such systems confined in QD by an order of magnitude in comparison with the energies values for same systems in bulk (Stébé *et al.*, 1989). This fact suggests that the confined systems are more stable avoiding their dissociation.

Recently, it has been reported the fabrication of self-assembled quantum rings (QRs) with a thickness of about 2 nm, outer radius between 30 and 70 nm and a well defined centre hole of about 10 nm radius (Lorke *et al.*, 2000). This finding has attracted a strong theoretical interest in the study of the QR few-particle spectrum. Exact series solution for a two-electron system in the extreme adiabatic limit has been analyzed (Zhu *et al.*, 2003) as the width and the height of the QR become negligible. With the purpose of taking into account the effect of the finite QR thickness, in Ref. (Betancur *et al.*, 2007) it has been recently used the adiabatic approximation. The authors calculated the low-lying states of an on-axis negatively charged donor in a toroidal-shaped quantum ring with cross-section of small diameter. By using a similar approach in the present work, it is studied the low-lying states of an artificial hydrogen molecule H_2 which consists of two on-axis ionized donors and two electrons furnished by them in a narrow two-dimensional ring. This model allows us to study the evolution of the artificial hydrogen molecule H_2 energy spectrum to the

artificial negative hydrogen ion spectrum analyzed in Ref. (Betancur *et al.*, 2007) by changing the position of one donor while the other one remains fixed.

1.1 Theoretical model

We consider an artificial hydrogen molecule formed by two on-axis donors located at the points $R_1=(0, 0, a)$ and $R_2=(0, 0, -b)$, respectively, and two electrons released by them inside of a narrow two-dimensional ring located on the X-Y plane (see fig. 1). The confinement potential is determined by a circular ring with outer and inner radii equal to R_{out} and R_{in} , respectively. In the analysis, it has been introduced the mean radius parameter R_a defined as the half of the sum of R_{in} and R_{out} , that is, $R_a=(R_{in} + R_{out})/2$. Additionally its thickness is given by $w=R_{out} - R_{in}$.

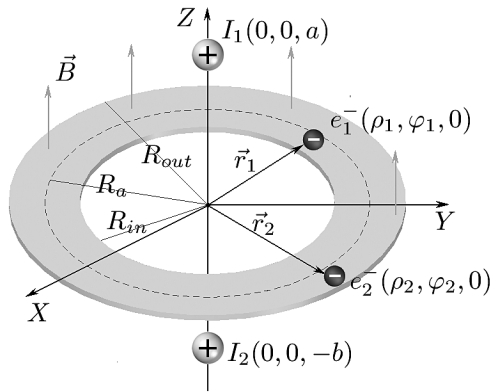


Figure 1. Artificial hydrogen molecule consisting of two electrons released by two on-axis impurities inside of a narrow two-dimensional nanoring. In this versatile model the impurities are located along the z-axis and their positions can be adjusted through the parameters a and b .

By using the effective-mass approximation, the dimensionless Hamiltonian of this system in presence of an external and homogeneous magnetic field $\mathbf{B} = B\hat{\mathbf{k}}$, can be explicitly written in cylindrical coordinates (ρ, φ, z) as:

$$H = \sum_{k=1}^2 \left[-\frac{\hbar^2}{2m} \nabla_{\mathbf{r}_k}^2 + V(\mathbf{r}_k) - \frac{2}{|\mathbf{r}_k - \mathbf{R}_1|} - \frac{2}{|\mathbf{r}_k - \mathbf{R}_2|} \right] + U_{ee} + U_{ii} \quad (1a)$$

$$U_{ee}(|\mathbf{r}_1 - \mathbf{r}_2|) = \frac{2}{|\mathbf{r}_1 - \mathbf{r}_2|} \quad U_{ii} = \frac{2}{|\mathbf{R}_1 - \mathbf{R}_2|} \quad (1b)$$

Where \mathbf{r}_k and \mathbf{R}_k denotes the electrons and ions positions vectors respectively, and $V(\mathbf{r}_k)$ is the confinement potential equal to zero and infinite inside and outside the

circular ring area, respectively. Remaining terms in the Hamiltonian (1) are the Coulomb interaction energies between electrons and ions (terms with minus sign) and the last two positive terms are the electron-electron (U_{ee}) and ion-ion repulsion (U_{ii}) respectively. With the purpose of comparing our results with those obtained for Zhu, 2003 y Betancur, 2007, we have used the effective Bohr radius $a_0^* = \hbar^2 \epsilon/m^* e^2$, the effective Rydberg $Ry^* = e^2/2\epsilon a_0^*$ and $\gamma = e\hbar B/2m^* c Ry^*$ as units of length, energy, and the conventional dimensionless magnetic field strength respectively. Since the two hydrogenic system described by the Hamiltonian (1) is not completely separable, its solution requires of an approximate method. Consequently, it was considered a narrow circular QR that satisfies the experimental condition ($R_a \gg w$) allowing us to decouple the fast radial electron motion from their rotational motion around the Z-axis. This separation can be achieved by using the well-known adiabatic procedure (Betancur *et al.*, 2007). In this sense, primarily it is necessary to find out the ground state wave function $f_0^{(k)}(\rho_k)$ ($k = 1,2$) and its corresponding energy E_0 for each electron into an infinite one-dimensional quantum well of length w . In second place, in order to obtain the all low lying-energy levels we have to solve the eigenvalue problem given by the effective Hamiltonian.

$$\hat{H}_a = -\frac{1}{R_a^2} \left(\frac{\partial^2}{\partial \varphi_1^2} + \frac{\partial^2}{\partial \varphi_2^2} \right) - i\gamma \left(\frac{\partial}{\partial \varphi_1} + \frac{\partial}{\partial \varphi_2} \right) + \frac{1}{4} \gamma^2 \langle \rho_1^2 + \rho_2^2 \rangle + 2E_0 + \frac{2}{(a+b)} + \bar{V}_{ee}(\varphi_1 - \varphi_2) + \bar{V}_{ei} \tag{2}$$

The mean effective potentials related to the electrons-ions attraction, \bar{V}_{ei} and the two-electron repulsion, $\bar{V}_{ee}(\varphi_1 - \varphi_2)$ are given by the following expressions:

$$\bar{V}_{ei} = -2 \left\langle f_0^{(k)} \left| \frac{1}{\sqrt{\rho_k^2 + a^2}} + \frac{1}{\sqrt{\rho_k^2 + b^2}} \right| f_0^{(k)} \right\rangle; k = 1,2 \tag{3a}$$

$$\bar{V}_{ee}(\varphi) = \left\langle f_0^{(1)} f_0^{(2)} \left| \frac{2}{\sqrt{(\rho_1 - \rho_2)^2 + 4R_a^2 \sin^2(\varphi/2)}} \right| f_0^{(1)} f_0^{(2)} \right\rangle \tag{3b}$$

$$\langle \rho_k^2 \rangle = \left\langle f_0^{(k)} \left| \rho_k^2 \right| f_0^{(k)} \right\rangle; k = 1,2 \tag{3c}$$

It is important to notice that the mean effective potential in the Hamiltonian (2) depends only on the difference of the angular coordinates, thus by using the centre of mass, $\theta = (\varphi_1 + \varphi_2)/2$ and relative, $\varphi = \varphi_1 - \varphi_2$ coordinates, the two-particle Hamiltonian (2) can be separated into the centre-of-mass \hat{H}_c and relative \hat{H}_r terms:

$$\hat{H} = \hat{H}_c + \hat{H}_r + 2E_0 + \bar{V}_{ei} + \frac{2}{(a+b)} \quad (4a)$$

$$\hat{H}_c = -\frac{1}{2R_a^2} \frac{d^2}{d\theta^2} - i\gamma \frac{d}{d\theta} + \frac{1}{4}\gamma^2 \langle \rho_1^2 + \rho_2^2 \rangle \quad (4b)$$

$$\hat{H}_r = -\frac{2}{R_a^2} \frac{d^2}{d\varphi^2} + \bar{V}_{ee}(\varphi) \quad (4c)$$

For comparison purposes, we use the same energy labelling notation $E(M, m, s)$ used in Ref. (Zhu *et al.*, 2003) (Betancur *et al.*, 2007) where it is explicitly written the three quantum numbers, the centre of mass angular momentum M , the relative angular momentum m , and the spin s . The eigen functions of the Hamiltonian (4a) can be written as:

$$\Psi(\varphi_1, \varphi_2) = e^{iM\theta} \Phi_{m,s}(\varphi); \quad M, m = 0, \pm 1, \pm 2 \dots; \quad s = 0, 1 \quad (5)$$

Here, the functions $e^{iM\theta}$ are the solutions of the eigenvalue problem given by the Hamiltonian \hat{H}_c and with exact eigen values

$$E_c(M) = M^2/2R_a^2 + M\gamma + \frac{1}{4}\gamma^2 \langle \rho_1^2 + \rho_2^2 \rangle \quad (6)$$

The remaining term $\Phi_{m,s}(\varphi)$ in (5) is the solution of the following one-dimensional wave equation corresponding to the relative angular motion

$$-\frac{2}{R_a^2} \frac{d^2 \Phi_{m,s}(\varphi)}{d\varphi^2} + \bar{V}_{ee}(\varphi) \Phi_{m,s}(\varphi) = E_r(m, s) \Phi_{m,s}(\varphi) \quad (7a)$$

$$\Phi_{m,s}(-\varphi) = (-1)^s \Phi_{m,s}(\varphi); \quad \Phi_{m,s}(\varphi)(-1)^M = \Phi_{m,s}(\varphi \pm 2\pi) \quad (7b)$$

Where $E_r(m, s)$ correspond to the eigen values of the \hat{H}_r operator.

The total energy levels $E(M, m, s) = E_c(M) + E_r(m, s) + \bar{V}_{ei} + \frac{2}{(a+b)}$ are referred to the non-interacting two-electron ground state energy in an infinite quantum well, therefore the $2E_0$ term has been removed from the total energy $E(M, m, s)$. In our numerical work we have solved the wave equation (7a) with periodic boundary conditions (7b) by using trigonometric sweep method (Betancur *et al.*, 1998).

3. RESULTS AND DISCUSSION

In figure 2 it is displayed the H_2 artificial molecule total energy for the some low-lying states at zero magnetic field, as a function of the b parameter fixing the location of the other impurity in $a = 20a_0^*$. The solid lines represent our results and the horizontal dotted lines are the energy values for a negative ion obtained in Ref. (Betancur *et al.*, 2007). As can be seen, our results consistently tend to those obtained for a negative ion as one of the impurities is moved systematically away along the symmetry z -axis while the other impurity remains fixed. In the inset we show an asymptotic tendency for very large values of b parameter corresponding to the energy values for a negative hydrogen ion D^- . The discrepancy in the limiting case, tend to be smaller than $0.02Ry^*$, but never equal to zero. This behavior is a consequence of the influence of the cross-section morphology on the energy spectrum, since in Ref. (Betancur *et al.*, 2007) was used a toroidal quantum ring to confine the two-electron system instead of our two-dimensional nanoring. In this sense, these facts are an indirect proof of the good accuracy and validity of our numerical results obtained in the present work.

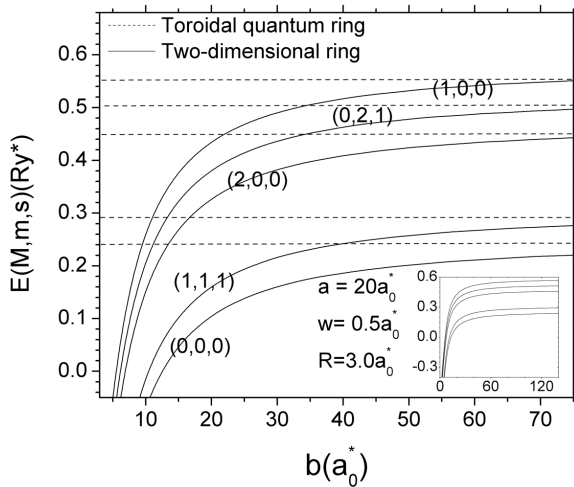


Figure 2. Total energy for some low-lying states as a function of the b parameter for a narrow two-dimensional ring of average radius equal to $3a_0^*$, and with a thickness of $0.5a_0^*$. Our results (solid lines) consistently tend to those obtained with Betancur *et al.*, 2007 for a negative ion in a toroidal ring-shaped (horizontal dashed lines).

The renormalized energies $\tilde{E} = ER_a^2$ as a function of the average radius for some low-lying levels are displayed in Fig. 3 (a). It is noticeable the changing of the renormalized energy when the average radius begins to increase from zero to $15a_0^*$ which it is

explained as an effect of the strong competition between the \bar{V}_{ee} , \bar{V}_{ei} and $2/(a+b)$ terms in Hamiltonian (2). For instance, the states (2,2,1) and (2,2,0), take positive values for average radii lower than $1a_0^*$ and greater than $16a_0^*$ owing to the small values of R_a the electron-electron and ion-ion repulsion predominates, but as the average radius is increased the electron-ion attraction overweighs. If the process of increasing R_a continues, there is a point where the \bar{V}_{ee} and $2/(a+b)$ becomes again predominant. The fig. 3(a) supports this idea since it clearly shows that the minima of the total mean effective potential (located at $\varphi = \pi$) takes positive value for $R_a = 1a_0^*$, then by increasing the average radius, the minima takes negative values up to $8a_0^*$ when the maximum negative value is reached. From this point the minima increases until approximately $13a_0^*$ when the minima takes a zero value, and subsequently the renormalized mean effective potential takes only positive values. This process is more easily seen in the inset of the fig. (3b) where it is plotted the minima potential $\bar{V}(\varphi = \pi)$ as a function of R_a .

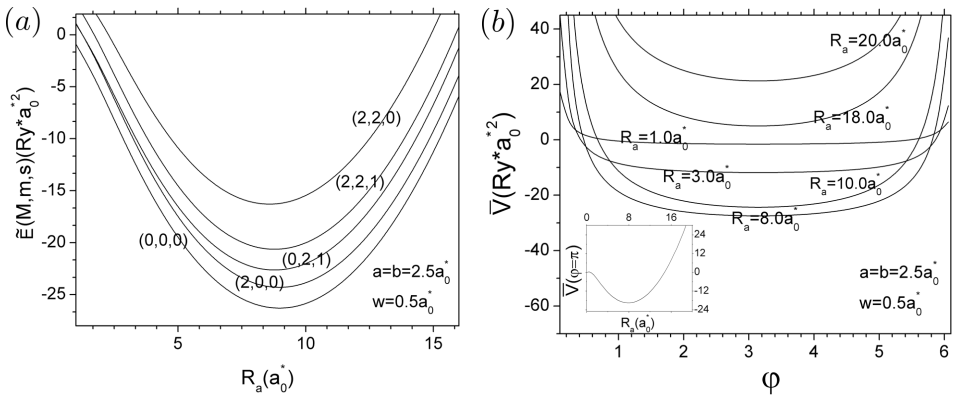


Figure 3. Renormalized energy as a function of average radius R_a for a symmetric configuration of two donors symmetrically located respect to the center of the ring for some low-lying levels (a) The total renormalized mean effective potential $\bar{V} = R_a^2(\bar{V}_{ee} + \bar{V}_{ei} + 2/(a+b))$ as a function of the φ relative coordinate (b).

In figs. (4a) and (4b) are shown the renormalized energies for large values of donor-donor separation. In particular, for $a = b = 50a_0^*$ is noticeable that for values of R_a greater than $18a_0^*$ the some low lying energy levels take negative values as an indicative of the predominance of the attractive interactions. In contrast, for larger values of donor-donor separation as can be seen in fig. (4b) ($a = b = 200a_0^*$) the attractive interaction becomes negligible, and therefore the donor-donor and electron-electron repulsive interactions overweighs.

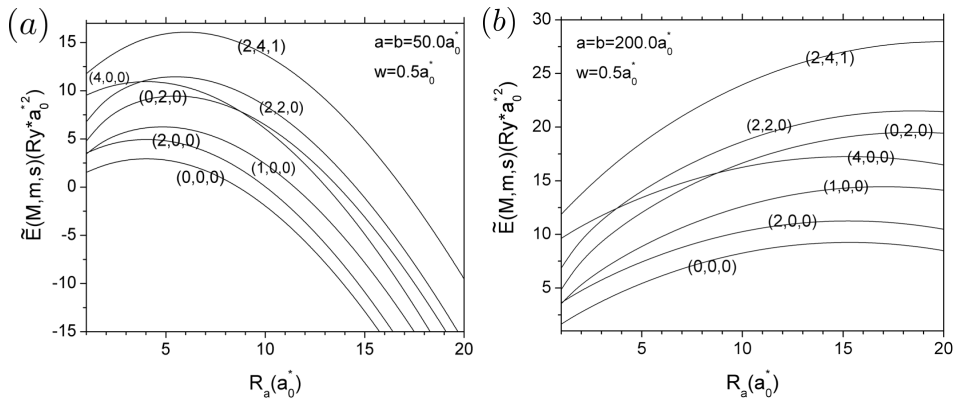


Figure 4. Renormalized energy as a function of R_a for impurities symmetrically placed respect to the origin, in the first case they are nearer with a donor-donor separation equal to $100 a_0^*$ (a) in the last case they are separated by a distance equal to $400 a_0^*$ (b).

In figs. (5a) and (5b) is shown the total energy as a function of the a parameter for two different values of b . The curves despite of exhibiting a similar behavior, the energy values are strongly dependent on the impurities location. These results illustrate a wide range of possibilities to modify the equilibrium distance (located near $5.0a_0^*$ and $0.0a_0^*$ for figures (5a) and (5b), respectively) and the barrier height (with values close to $-1.16Ry^*$ and $-0.92a_0^*$ for figures (5a) and (5b), respectively) of the artificial molecule by varying the impurities location along the symmetry axis.

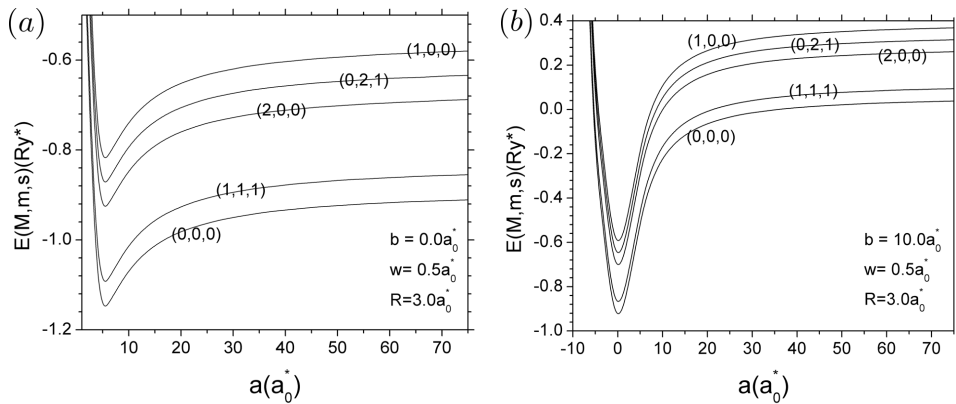


Figure 5. Total Energy as a function of a parameter for values of $b = 0.0a_0^*$ (a) and $b = 10.0a_0^*$ by varying the impurities location along the symmetry axis is possible to tailor the energy spectrum of this artificial H_2 molecule since it modifies the equilibrium distance and the barrier height.

In figs. (6a) and (6b) is displayed the renormalized energy as a function of the of the dimensionless magnetic field strength parameter γ . In order to understand this remarkable evolution of the energy curves, one must keep in mind that these curves are the result of the strong competition between two pairs of parameters present in the Hamiltonian (2). The first pair corresponds to the repulsive and attractive potential terms and second one corresponds to the paramagnetic and diamagnetic terms. The first pair of parameters allows us to explain the reason for which the energy takes positive or negative values. For instance, if the separation between ions is smaller than $2.5a_0^*$ the contribution of the ion-electron attraction term is predominant and the total energy takes negative values (fig. 4b) but for larger separations, the repulsive term becomes more significant and the total energy take positive values (fig. 6a).

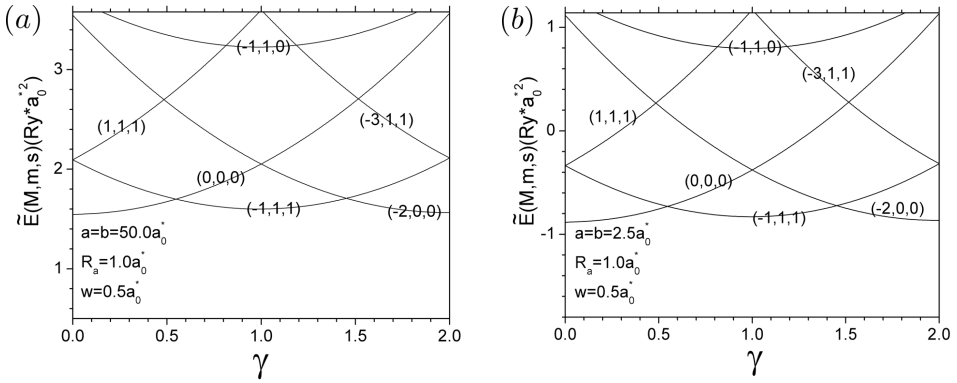


Figure 6. Total Energy as a function of the conventional dimensionless magnetic field strength parameter γ for values of $a = b = 50.0a_0^*$ (a) and $a = b = 2.5a_0^*$ (b).

The presence of the magnetic field in the second pair of parameters allows us to explain two important aspects. On one hand, due to the paramagnetic term varies linearly with the magnetic field and the angular momentum M , this term is responsible of the splitting in the energy levels whose angular momentum M has different signs. For instance, when $M \geq 0$ all curves are linear at beginning with positive slope but when $M < 0$ the slope becomes negative. On the other hand, and according to the figures 6 we can observe an oscillatory behavior in the molecular complex ground state energy with period equal to one. This result corresponds to the well known Aharonov-Bohm effect (Aharonov *et al.*, 2004). In order to understand the behavior of these curves is necessary to take into account that for small values of the magnetic field strength the paramagnetic term predominates on the diamagnetic term which is quadratic with the magnetic field. Nevertheless, this behavior changes for larger


values of the magnetic field strength since the contribution of the diamagnetic term to the total energy becomes more significant than the paramagnetic term which allows us to explain the parabolic-shape of the all curves from a given value of the field strength.

In short, our model of an artificial hydrogen molecule consisting of two electrons released by two on-axis donors inside of a two-dimensional narrow ring allows us to explore a great variety of properties, such as the transition to a negative ion when one of the donors is moved away large distances from the another impurity assumed fixed. Additionally the shapes and positions of the potential curves in the artificial hydrogen molecule may be tailored by the impurities location.

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