

Effect of Temperature and Pressure on Correlation Energy in a Triplet State of a Two Electron Spherical Quantum Dot

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Abstract

The combined effect of hydrostatic pressure and temperature on correlation energy in a triplet state of two electron spherical quantum dot with square well potential is computed. The result is presented taking GaAs dot as an example. Our result shows the correlation energies are i) negative in the triplet state contrast to the singlet state ii) it increases with increase in pressure iii) further decreases due to the application of temperature iv) it approaches zero as dot size approaches infinity and v) it contribute 10% decrement in total confined energy to the narrow dots. All the calculations have been carried out with finite models and the results are compared with existing literature.

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1. Introduction

The development of low dimensional semiconductor system (LDSS) has found wider application in many areas of nanoscience nowadays [1, 2]. Semiconductor quantum dot (QD) is an excellent system to investigate the confinement effects [3, 4]. The fabrication of semiconductor quantum structures in quasi zero dimension will show exotic electron behavior due to electronic confinement [5]. The energy levels of the confined electron in a QD are discrete as in an atom [6, 7]. Hence QD is called as artificial atoms [8]. The confinement will affect due to correlation energy

(CE). The confinement increases in the singlet state [9] and it decreases in the triplet state due to the presence of CE [10]. The correlation effects were treated to various degrees of theoretical sophistication by several authors [11,12]. The CE (electron-electron coulomb interaction energy) plays a vital role in varying the physical properties of LDSS such as optical and transport phenomena. Hence the interaction in a QD is especially interesting.

The combined effect of hydrostatic pressure and temperature on the optical and electrical properties in LDSS has been investigated by several authors

[13-25]. Kirak et al. [13] studied the effects of the hydrostatic pressure and temperature on the binding energy and nonlinear optical properties for a SQD with parabolic confinement. He found that the binding energy increases by increasing pressure and decreases by temperature. A.M. Elabasy [14, 15] founded that the binding energy of the donor electron, associated with the donor ion, decreases with enhancing the temperature. Karimi et al. [16] indicate that the hydrogenic impurity, hydrostatic pressure and temperature increase the energy difference between the sub-bands in a multilayered SQD. Kospoglu et al. [17] found that an increment in temperature results in a decrement in donor impurity binding energy while an increment in the pressure for the same temperature enhances the binding energy in a cylindrical quantum wire. Yesilgul et al. found that the photo ionization cross-section increased as temperature increased. Conversely, hydrostatic pressure decreased, the photo ionization cross-section increased on a QD [18] and Quantum well [19]. Safarpour et al. [20] found that the electronic energy levels in a SQD increases as the pressure increases and decreases as the temperature increases. Elmeshad et al. [21] studied the effects of the hydrostatic pressure and temperature on the exciton binding energy confined inside a cylindrical GaAs QD. Rezaei et al. found that hydrostatic pressure and temperature have a great influence on the binding energy in a two-dimensional QD [22] and spherical Gaussian QD [23].

In the above all investigation, the effect of temperature and pressure on the CE in a SQD is neglected. Rejo Jeice et al. [10] studied the CE in a triplet of a two electron SQD. Sivakami et al. [24] studied the effect of hydrostatic pressure and temperature on CE in the finite barrier model. The calculations were restricted to the singlet state. More over in the previous paper one of the authors

had studied the effect of hydrostatic pressure and polaronic mass on the CE in a SQD [25].

The purpose of the present work is to extend the calculation of Ref. [10] to the (1s-1p) triplet state in a two electron SQD and to investigate theoretically the effects of hydrostatic pressure and temperature on it. The plan of the present work is as follows. In section 2 we present the mathematical models and details of calculations. The results obtained are presented in section 3 with a detailed discussion of the results. Conclusions are presented in section 4.

2. Models and calculations

2.1. Two electrons in a spherical quantum dot

Here we consider two electrons in a Spherical quantum dot (SQD) in the finite barrier model. In the effective mass approximation, the hydrostatic pressure and temperature dependent Hamiltonian is given by

$$H = \frac{P_1^2}{2m^*(P,T)} + \frac{P_2^2}{2m^*(P,T)} + V_D(\vec{r}_1, \vec{P}, T) + V_D(\vec{r}_2, \vec{P}, T) + \frac{e^2}{\epsilon_{w,b}(P,T) |\vec{r}_1 - \vec{r}_2|} \quad (1)$$

We write $H = H_1 + H_2 + H^l$ with

$$H^l = \frac{e^2}{\epsilon_{w,b}(P,T) |\vec{r}_1 - \vec{r}_2|}$$

where, $m^*(P,T)$, $\epsilon_{w,b}(P,T)$, $V_D(\vec{r}_i, P, T)$ are the hydrostatic pressure and temperature dependent effective-mass, dielectric constant and confining potential, respectively. The two subscripts d and b stand for the potential dot (GaAs) and the potential barrier ($\text{Ga}_{1-x}\text{Al}_x\text{As}$), respectively. The values of $m^*(P,T)$, $\epsilon_{w,b}(P,T)$, are taken from Refs. [15, 26] and barrier height values are given in Table 1. P is the hydrostatic pressure in GPa and T is the absolute temperature in Kelvin. In Hamiltonian (1), r_1 , r_2 denote the electron positions inside the dot of radius $R(P,T)$, which also depend on pressure and

temperature. In our numerical calculations, we use atomic units in which $m_0 = e^2 = \hbar^2 = 1$. The confining potential is given by,

$$V_D(r_i, P, T) = \begin{cases} 0 & r_1 r_2 \leq R \\ V_0 & r_1 r_2 \geq R \end{cases} \quad (2)$$

where $V_0 = Q_c \Delta E_g(x, P, T)$ is the barrier height, Q_c is the conduction band offset parameter which is taken to be 0.6 [27]. The band gap difference depends of the concentration of Al. In our case $Ga_{1-x}Al_xAs$ is the barrier medium in which GaAs dot is embedded. The total energy band gap difference [1] between the GaAs dot and the $Ga_{1-x}Al_xAs$ barrier media, as a function of x , is given by

$$\Delta E_g(x) = 1.155x + 0.37x^2 \text{ eV} \quad (3)$$

In the present work, we have chosen $x = 0.3$ and 0.4 and the value of V_0 turns to be 227.88 meV and 312.72 meV respectively. The eigen functions for the two lowest lying states within the dot is given by [6]

$$\psi_{1s}(\vec{r}_i) = \begin{cases} N_1 \frac{\sin(\alpha_i r_i)}{\alpha_i r_i} & r_i \leq R \\ A_1 \frac{e^{-\beta_i r_i}}{\beta_i r_i} & r_i \geq R \end{cases} \quad (4)$$

$$\psi_{1p}(\vec{r}_i) = \begin{cases} N_2 \left[\frac{\sin(\alpha_i r_i)}{(\alpha_i r_i)^2} - \frac{\cos(\alpha_i r_i)}{\alpha_i r_i} \right] \cos \theta & r_i \leq R \\ \int \psi_A^*(\vec{r}_1, \vec{r}_2) \psi_A(\vec{r}_1, \vec{r}_2) d\tau_1 d\tau_2 & (5) \end{cases}$$

$$iA_2 \left[\frac{1}{\beta_i r_i} + \frac{1}{(\beta_i r_i)} \right] e^{-\beta_i r_i} \cos \theta \quad r_i \geq R$$

where N_1, N_2, A_1 and A_2 are normalization constants and α_i and β_i are given

$$\alpha_i = \sqrt{2m^*(P, T)E(P, T)} \quad \text{and} \\ \beta_i = \sqrt{2m^*(V_0(P, T) - E(P, T))}$$

Matching the wave function and their derivatives at the boundary and imposing the Ben Daniel and Duke boundary condition [28] the 1s-state and 1p-state energies are obtained. Using the E_{1s} and E_{1p} state energies the triplet state confined energies (Binding energy) and the CE is found out.

In the two-electron system we calculate the CE using the perturbation method. Here H^1 is the perturbation term and ϵ_0 is the static dielectric constant. In our problem we have considered one electron in the ground state (i.e. 1s-state) and another in the excited state (1p-state). Therefore we may obtain a triplet state and the total spin of the system is 1. In this situation the wave function of the triplet state is [9].

$$\psi_A(\vec{r}_1, \vec{r}_2) = \frac{1}{\sqrt{2}} [\psi_{1s}(\vec{r}_1)\psi_{1p}(\vec{r}_2) - \psi_{1s}(\vec{r}_2)\psi_{1p}(\vec{r}_1)]$$

This is spatially anti-symmetric. In Eq. (6) $\psi_{1s}(r_i)$ and $\psi_{1p}(r_i)$ are (with $i=1, 2$) as given in equations (4) and (5).

$$= \left(\frac{3R}{2\alpha_1^2} - \frac{3\sin(2\alpha_1 R)}{4\alpha_1^3} - \frac{3 \cdot e^{-2\beta_1 R}}{2\beta_1^3} \right) * \left(\frac{2}{R^2} \left(\frac{R}{2\alpha_2^4} - \frac{\sin(2\alpha_2 R)}{4\alpha_2^5} \right) - \frac{\sqrt{2}}{R} \left(\frac{1 - \cos(2\alpha_2 R)}{2\alpha_2^4} \right) + \frac{R}{2\alpha_2^2} \right) \\ + \frac{\sin(2\alpha_2 R)}{4\alpha_2^3} + \frac{e^{-2\beta_2 R}}{\beta_2^3} \left(\frac{1}{2} + \frac{1}{\sqrt{5}\beta_2 R} + \frac{1}{10\beta_2^2 R^2} \right) \quad (6)$$

We obtain the correlation energy for the triplet state is given by

$$\Delta E = \int \psi_A^*(\vec{r}_1, \vec{r}_2) \frac{e^2}{\epsilon_0 |\vec{r}_1 - \vec{r}_2|} \psi_A(\vec{r}_1, \vec{r}_2) d\vec{r}_1 d\vec{r}_2 \quad (7)$$

where $\psi_A(\vec{r}_1, \vec{r}_2)$ is given in Eq.(6). In order to evaluate some of the integrals we use mean value theorem. After a lengthy, but straight forward algebra we obtain ΔE . The evaluation of ΔE (first order correction) is tedious for triplet state and the expression is lengthy. Hence we refrain from giving it here. However the evaluation of is given in Ref [10] and the results obtained are given in table 2 and figure 1-3.

2.2. Effect of Temperature and Pressure

The hydrostatic pressure and temperature dependent conduction band effective mass of GaAs and $\text{Ga}_{1-x}\text{Al}_x\text{As}$ can be written as [29,30]

$$\frac{m_e}{m^*(P,T)} = 1 + E \left\{ \frac{2}{E_g^\Gamma(P,T)} + (E_g^\Gamma(P,T) + \Delta_0)^{-1} \right\}$$

$$12.74 \exp(-1.73 \times 10^{-3} P) \exp[9.4 \times 10^{-5} (T - 75.6)]$$

$$T \leq 200\text{K}$$

$$\epsilon_{d,b}(P,T) \quad (11)$$

$$13.18 \exp(-1.73 \times 10^{-3} P) \exp[20.4 \times 10^{-5} (T - 300)]$$

$$T \geq 200\text{K}.$$

The corresponding dielectric constant of $\text{Ga}_{1-x}\text{Al}_x\text{As}$ is given as $\epsilon_b(P,T) = \epsilon_d(P,T) - 3.12x$

$$(12)$$

From equ.(2) the pressure and temperature dependent barrier height, the band gap difference is given by

$$\Delta E_g(x,P,T) = \Delta E_g(x) + D(x)P + G(x)T \quad (13)$$

where $D(x) = [-1.3 \times 10^{-3}]x$ eV/kbar and $G(x) = [-1.15 \times 10^{-4}]x$ eV/K. The variation of dot size with pressure is given [35] by

$$R(P) = R_0(1 - 1.5082 \times 10^{-3} P) \quad (14)$$

Where $E = 7.51\text{eV}$, is the energy related to the momentum matrix element. $\Delta_0 = 0.341\text{ eV}$ is the spin-orbit splitting, m_e is the free electron mass and $E_g^\Gamma(P,T)$ is the pressure and temperature dependent energy gap for the GaAs QD at the τ -point and is given by [31,32]

$$E_g^\Gamma(P,T) = E_g^\Gamma(0,T) + bP + cP^2 \quad (8) \quad \text{where}$$

$$b = 1.26 \times 10^{-2} \text{eVbar}^{-1}, \quad c = 3.77 \times 10^{-5} \text{eVbar}^{-2} \quad \text{and}$$

$$E_g^\Gamma(0,T) = 1.519 - \frac{(5.405 \times 10^{-4} T^2)}{(T + 204)} \quad (9)$$

The effective mass of $\text{Ga}_{1-x}\text{Al}_x\text{As}$ is given by [33]

$$m_b^*(P,T) = m_d^*(PT) + 0.083x \quad (10)$$

where x is the aluminum composition. The variation of dielectric constant with pressure and temperature is given as [34],

In the numerical work the pressure used was 0-4 GPa, which corresponds to 40 kbar. We have not considered pressures beyond 4 GPa, because of a direct to indirect band gap transition of GaAs at about 4 GPa [36]. Also the aluminum concentration is also $x \leq 0.4$. Since the indirect band gap nature in $\text{Ga}_{1-x}\text{Al}_x\text{As}$.

3. Result and Discussion

The results obtained are shown in Tables 1-2 and Figs. 1-5. We have computed the combined effect of hydrostatic pressure and temperature effects on the SQD. The various parameters used in the

calculation and their values at various temperatures are given in Table 1.

Table 1. Variation of the effective mass and dielectric constant for the GaAs and Ga_{1-x}Al_xAS at aluminium composition, x = 0.4.

T (K)	Dielectric constant				Barrier height (meV)
	Effective mass				
	GaAs	Ga _{1-x} Al _x	GaAs	Ga _{1-x} Al _x	
0	0.067	0.100	12.65	11.402	312.72
100	0.066	0.100	12.77	11.521	309.96

200	0.064	0.098	12.91	11.642	307.20
300	0.063	0.096	13.18	11.764	304.44
400	0.061	0.095	13.45	11.886	301.68
500	0.059	0.093	13.73	12.011	298.92
600	0.057	0.091	14.01	12.136	296.16

From it, we find that the increase in temperature will decrease the values of both the effective mass and the barrier height and increase the value of dielectric constant of GaAs and Ga_{1-x}Al_xAs.

Table 2. Temperature and pressure dependent correlation energies in a spherical quantum dot.

Pressure (GPa)	Correlation Energy (meV)							
	T=0K				T=600K			
	R=50Å	R=65Å	R=100Å	R=150Å	R=50Å	R=65Å	R=100Å	R=150Å
0	-43.99	-24.56	-15.09	-9.63	-40.02	-22.24	-13.99	-8.79
1	-44.50	-24.84	-15.17	-9.69	-41.13	-21.34	-14.37	-8.94
2	-44.94	-25.57	-15.27	-9.77	-41.79	-23.45	-14.65	-9.38
3	-45.67	-25.71	-15.36	-9.89	-42.67	-23.98	-14.93	-9.65
4	-46.05	-25.97	-15.57	-9.92	-43.57	-24.65	-15.38	-9.97

Table 2 we have presented the temperature and pressure dependent correlation energies in a SQD. We notice that the correlation energy decreases as the dot size increases, a feature that is well known in literature [9,10]. The CE decreases as the temperature increases and it increases as the pressure increases. Also we observe that the CE is negative as expected for the triplet state. This is a consequence of the exchange interaction in the

triplet state. This interaction arises because of the anti-symmetric nature of the wave functions equ. (6). Fig.1 shows the variation of CE with different dot radius for different temperatures T=300 K and 600 K without the pressure. For the zero pressure (or constant pressure) we found that the C.E decreases for all the dot radii. It also decreases due to increase in temperature since the values of the effective mass decreases.

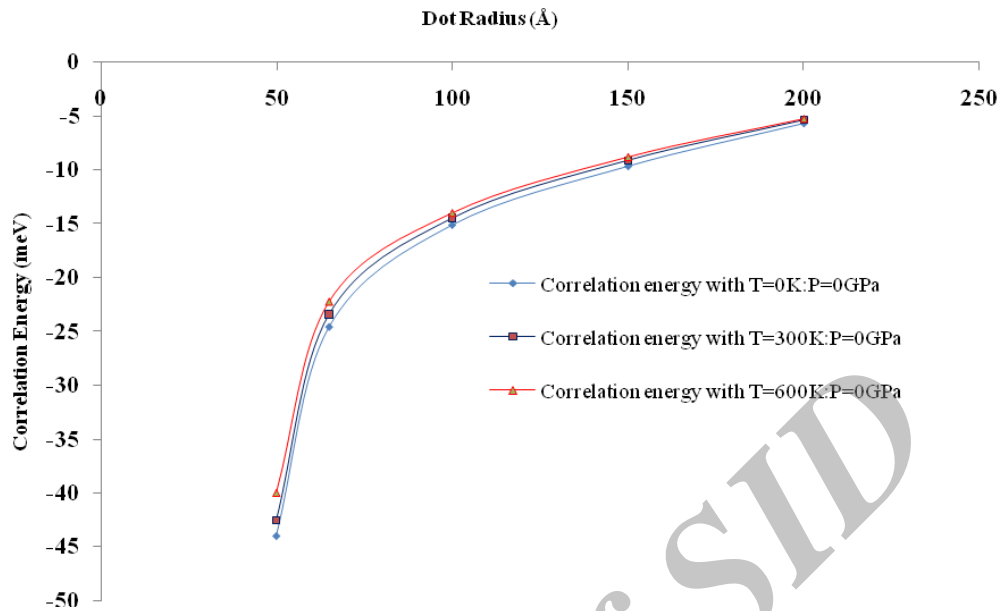


Fig. 1. Variation of correlation energy with different dot radius for different temperatures $T=300$ K and 600 K without the pressure when $x=0.4$

As the result the effective Bohr radius increases causing the potential barrier height to decrease. For larger dot size it shows 3D behaviour. For the smaller dot radii there is 5% decrease in correlation due to application of temperature. We found that the CE is higher for smaller dot size of higher aluminum concentration. It approaches to zero as the dot radius tends to infinity. Also we found that there is no CE for dot radii less than 50\AA , because no bound $1p$ state is possible since the barrier height itself is 312.72 meV when $x=0.4$.

Fig.2 shows the variation of CE with different dot radius for the temperature $T=600$ K with and without the pressure. We notice that the hydrostatic pressure increases the CE increases for all the dot sizes for the given temperature. The physical reason of this behavior is that as the pressure goes up, the wave functions are very confined for the dot and the effective mass and dielectric constant increases.

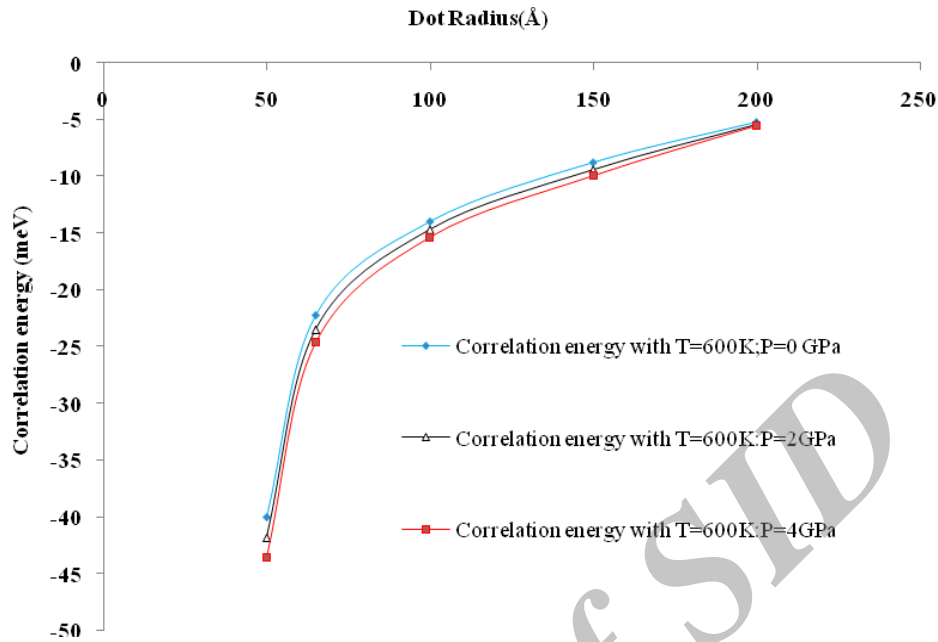


Fig. 2. Variation of correlation energy with different dot radius for the temperature $T = 600$ K with and without the pressure when $x=0.4$.

For the smaller dot radii there is 4.5% increase in correlation due to application of pressure. Thus the simultaneous application of pressure and temperature, it is observed that the CE decreases for all dot sizes. Fig.3 presents the variation of CE with different dot radius for the temperature $T=600$ K and $P=0$ GPa for the singlet and triplet state. Here we have considered the value of aluminum concentration as $x=0.3$ in order to compare with the reference singlet state energies [24]. We observe

that the triplet state energies is negative and it is almost equal to that of singlet state energies which is positive. In the singlet state there is no exchange interaction among the electrons of opposite spin orientations. Thus the CE is repulsive. However in the triplet state the exchange interaction is attractive which favors' the parallel arrangement of spins via Hund's rule as in ferromagnetism.

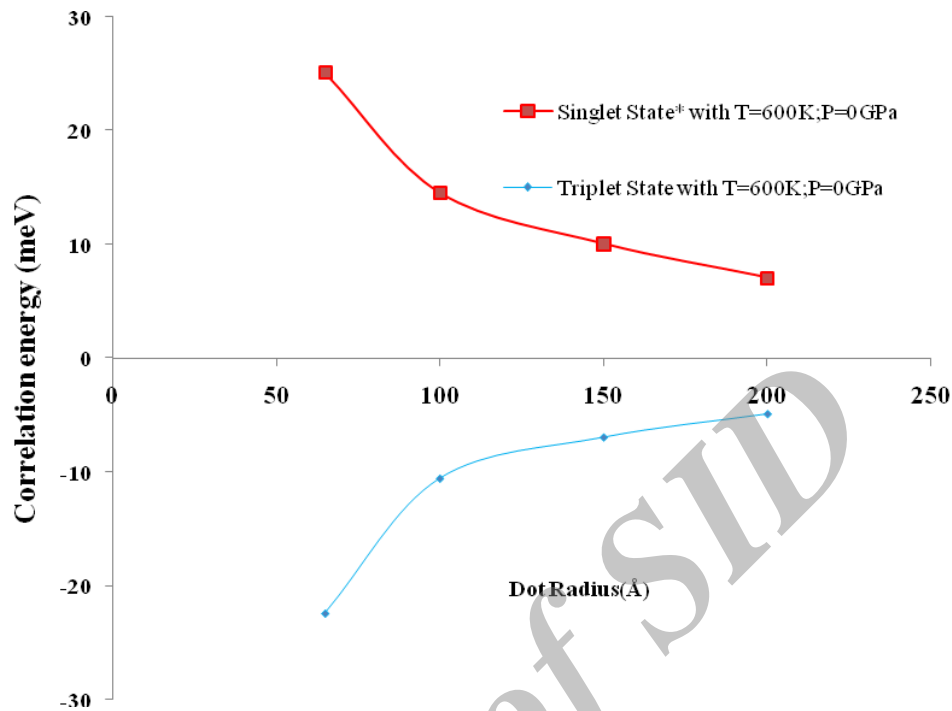


Fig. 3. Variation of correlation energy with different dot radius for the temperature $T=600$ K and $P=0$ GPa when $x=0.3$ (*Ref. [9])

There is no CE for to triplet state for the dot radii less than 65Å , because no bound $1p$ state is possible since the barrier height itself is 227.88 meV when $x=0.3$.

In Fig.4 we have presented the variation of CE with the applied pressure at different temperatures ($T=300$ and 600 K) and dot radii ($R=50$ and 100 Å). The correlation energies linearly increases with pressure. This is due to increase in effective mass and dielectric constant and barrier height. For smaller dot radius the variation of

correlation energy is higher as compare to larger dot radii.

In Fig.5 we display the variation of CE as a function of temperature with different pressures ($P=0$ and 4 GPa) and dot radii ($R=50$ and 100 Å). The correlation energy linearly decreases with temperature.

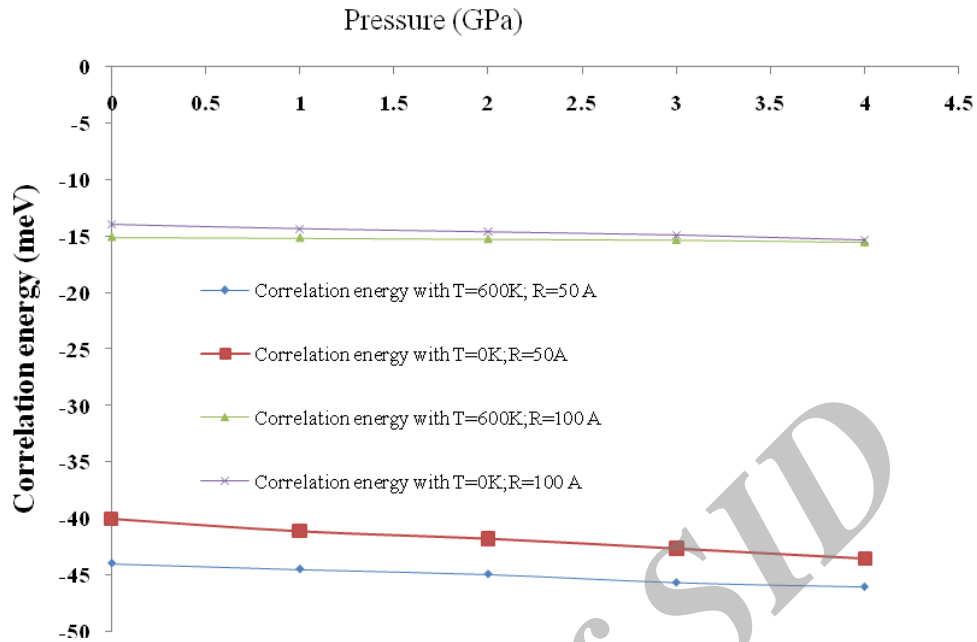


Fig. 4. Variation of correlation energy versus pressure with different temperatures ($T=300$ and 600 K) and dot radii ($R=50$ and 100 Å).

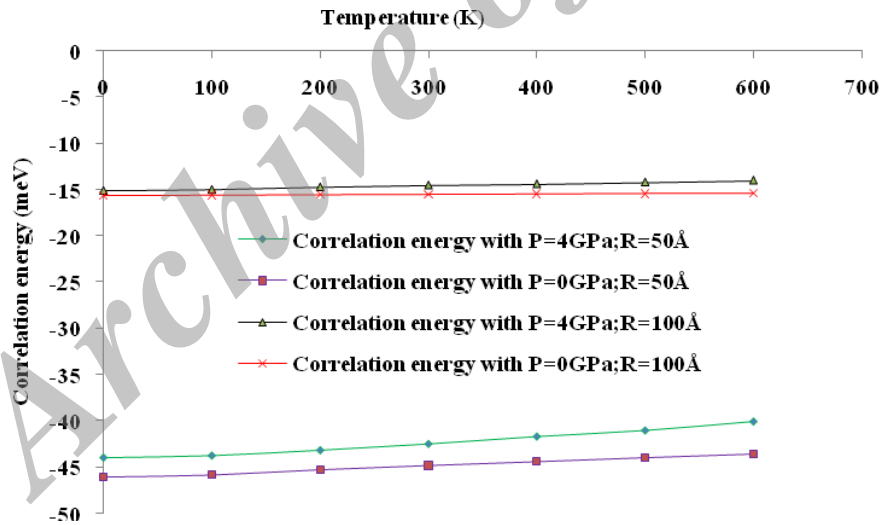


Fig. 5. Variation of correlation energy as a function of temperature with different pressures ($P=0$ and 4 GPa) and dot radii ($R=50$ Å and 100 Å).

This is due to increases in effective mass, dielectric constant and barrier height. Liang et al. found the ground state binding energy of the neutral

donor in a QD increased as hydrostatic pressure increased [37]. The physical origin of the result is related to the effect of the existence of the Coulomb

repulsion in an exciton–donor complex system. Moreover, we observed that the influence of pressure on the correlation energy is more obvious than temperature which is also due to columbic repulsion. The important conclusion that emerges from the result of Table 2 and Figures 1-5 is that the pressure and temperature effects are important for smaller dots and should be considered in the studies of LDSS.

4. Conclusion

In the present study we investigated the effects of hydrostatic pressure and temperature on the CE in the triplet state of GaAs/Ga_{1-x}Al_xAs for a two electron SQD by using a perturbation approach within the effective mass approximation. The important conclusions that emerge are i) the correlation energies are important for smaller dot size. ii) it is negative for triplet state and it contributes 10% decrement in total confined energy to the narrow dots. iii) the effect of hydrostatic pressure increase the correlation effects may be used to tune the output of optoelectronic devices without modifying the physical size of the quantum dot and iv) the effect of high temperatures is quite significant in small dot radii due to thermal broadening in the mixed quantum state.

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