



The Effect of Relativistic Quantum Corrections on the Thermal Properties of Three-Dimensional Spherical Semiconductor Quantum Dot Under a Magnetic Field

Research Article

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Abstract. The ground state and excited states energies of 2-electron quantum dot with spherical harmonic oscillator type potential has been determined. The energy spectrum and wave functions for the quantum dot of asymmetric confinement are obtained by analytically solving the eigenvalue equation in the magnetic field. The effect of various relativistic corrections to kinetic energy, Darwin term and spin-orbit for the zero-dimensional structure to the energy eigenvalues and wave functions is also investigated. The thermal properties like internal energy, entropy and free energy are discussed graphically with radius of quantum dot and pressure and are found to have interesting dependence on the radius and pressure parameter with relativistic corrections for our model.

Keywords. Quantum dot; Magnetic field; Relativistic corrections; Pressure; Thermodynamic properties

PACS. 73.21La, 65.80.-g, 71.70.Ej

Received: May 5, 2018

Accepted: May 17, 2018

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1. Introduction

The research on the low-dimensional structures has drawn considerable interest due to their unique properties [1] and potential applications in physics, chemistry and engineering, specifically in the development of semiconductor microelectronic and optoelectronic devices [2]

such as electro-optic modulators [3], inter-band lasers and inter-sub band long wavelength detectors [4]. These structures can be quantum dots, quantum wires, quantum wells, quantum rings, super-lattices, etc. [5–7]. Quantum dots, in particular, have extensively been explored both experimentally and theoretically in the last few years, among all the low-dimensional semiconductor structures [8, 10, 11]. The motion of electrons in quantum dots can be confined in all three dimensions with advanced manufacturing techniques thereby producing the zero-dimensional structures. The confinement of charge carriers in quantum dots or artificial atoms generates discrete energy levels with spacing of a few MeV [12]. Studies have shown that the confinement potential plays a very crucial role in determining the linear and non-linear optical, electrical and magnetic properties of quantum dots and is modulated for a wide range of dot sizes and shapes [13–22]. Also, these novel properties and its effect on impurity states are extensively investigated under the influence of external perturbations such as electron-phonon interaction, magnetic field, electric field, temperature, impurity and pressure [23–27]. Various models have been studied to understand the effect of confinement potential on dot parameters namely disk-like (cylindrical) model, rectangular confinement potential, spherical harmonic potential, non-spherical oscillator, ring shaped oscillator, ring shaped non-spherical oscillator [28–30].

In this work, we have studied the effect of relativity on the thermodynamic properties in the GaAs quantum dots. With an appropriate confining potential, the theoretical physics associated with the quantum dots is mainly concerned for the electron-electron interaction in a correct way at nanoscale thus the thermodynamic properties of nanostructures become an important subject in physical science. The energy levels and the modified wave functions for both the ground ($1s$) and first excited ($1p$) states in the presence of magnetic field and relativistic effects as perturbation terms were computed theoretically within the effective mass approximation. The effect of relativistic corrections on the thermodynamic parameters have been calculated as a function of dot radius and the pressure. Among the studies available on various physical properties of quantum dots, there are few studies on thermodynamics properties of quantum dots [31–36]. To the best of our knowledge, no investigation has so far been made to understand the dependence of internal energy, entropy and free energy on the dot radius and pressure with relativistic corrections to the kinetic energy, Darwin term and spin-orbit interactions.

The paper is organized as follows. In Section 2 we first give a theoretical background on the problem considered. The main idea of our approach is then summarized in the same section. In the same section the Relativistic corrections to the energy states are described. Section 3 describes the various thermodynamic properties mathematically. In Section 4 is dedicated to results of the calculations and their probable physical reasons are discussed and finally, the paper ends with a brief summary and concluding remarks.

2. Theory and Model

2.1 Energy Levels and Wave Functions

Here we are considering a two-electron spherical Quantum dot in the presence of magnetic field under the confinement spherical harmonic oscillator potential for GaAs quantum dot is model ledas:

$$V(r_i) = -V_0 e^{\frac{-r_i^2}{2R_P^2}} \quad (2.1)$$

where $V(r_i)$ is the confinement potential of the quantum dot, r_i is the position coordinate of the i th particle, V_0 is the depth potential and RP give the measure of Range of confinement potential which represents the size of quantum dot and P represents the hydrostatic pressure.

Under the approximation $r_i \ll R$, the unperturbed Hamiltonian for a system of two interacting electrons, H_0 reduces to

$$H_0 = -\frac{\hbar^2}{2m_p^*} \sum_{i=1}^N \nabla_{r_i}^2 + \gamma^2 \sum_i^N r_i^2 \tag{2.2}$$

where $\gamma^2 = \frac{V_0}{2R_p^2}$ and m_p^* is the effective electronic mass as a function of hydrostatic pressure.

To include the effects from the underlying lattice and the interaction with the electrons from the valence and core bands the effective mass approximation is taken into account. Thus, thenon-relativistic Hamiltonian of a system of 2 electrons in a spherical quantum dot under the electric and magnetic fields (2.2) can be written as:

$$H_T = H_0 + H'_{pot} + H'_{M1} + H'_{M2} + H'_{M3}, \tag{2.3}$$

$$H'_{pot} = \sum_{i=1}^N -\frac{e^2}{\epsilon_P r_i} - NV_0. \tag{2.4}$$

Here 1st two terms i.e. $H_0 + H'_{pot}$ represent the contribution of hydrogenic Impurity and ϵ_P represents the dielectric constant of impurity which depends on hydrostatic pressure and temperature as,

$$\epsilon_{P,T} = 12.74 * e^{-1.67*10^{-3}P} * e^{9.4*10^{-5}(T-75.6)}. \tag{2.5}$$

The third term represents the energy due to the interaction between particles orbital magnetic dipole moment $\left(\frac{qL}{2cm_p}\right)$ and magnetic field B .

$$H'_{M1} = -\frac{qBL \cos \theta \cos \varphi}{2m_p^* c}. \tag{2.6}$$

The fourth term gives ratio of the paramagnetic and diamagnetic contribution which is very small and thus can be ignored i.e.

$$H'_{M2} = \sum_{i=1}^N \frac{q^2 |B|^2 r_i^2 \sin^2 \theta}{8m_p^* c^2}, \tag{2.7}$$

where $\langle r_i^2 \rangle = a_0^2$ where a_0 is the Bohr radius, and shift due to Magnetic energy correction is given by the Paschen Back effect as

$$H'_{M3} = \left\langle \Psi_{nlm} \left| \frac{eBLiZ}{2m_p^* c} \right| \Psi_{nlm} \right\rangle = \Delta E_B = B \mu_B \tag{2.8}$$

and $\mu_B = \frac{e\hbar}{2m_p^* c}$ is Bohr's magneton.

The Schrodinger equation with spherical symmetric harmonic oscillator potential (2.4) is exactly solvable with the following Energy spectrum:

$$E_{n_i l_i} = \sum_{i=1}^N \left(2n_i + l_i + \frac{3}{2} \right) \hbar \omega \tag{2.9}$$

where $n_i l_i = 0, \pm 1, \pm 2, \dots$

The Eigen values ET for the Hamiltonian (2.5) is given by the Schrodinger equation:

$$H_T \Psi_{nlm} = E_T \Psi_{nlm} \quad (2.10)$$

and the solution of the unperturbed Hamiltonian is given by

$$H_0 \Psi_{nlm} = E_0 \Psi_{nlm} \quad (2.11)$$

where nlm are the atomic quantum numbers of the wave function $\Psi(r, \theta, \phi)$.

For any arbitrary state, the complete wave function, $\Psi(r, \theta, \phi)$, can be written as

$$\Psi(r, \theta, \phi) = \sum_{nl} N_{nl} R_{nl}(r) Y_l^m(\theta, \phi) \quad (2.12)$$

where spherical harmonic $Y_l^m(\theta, \phi)$ is the eigenfunction of $L^2(\theta, \phi)$ satisfying

$$L^2(\theta, \phi) Y_l^m(\theta, \phi) = l(l+1) \hbar^2 Y_l^m(\theta, \phi) \quad (2.13)$$

and the radial wave function $R_{nl}(r)$ is the solution of the equation

$$\left(\frac{d^2}{dr^2} + 2r \frac{d}{dr} - \frac{l(l+1)}{r^2} \right) R_{nl}(r) + \frac{8\pi^2 m}{\hbar^2} [E_{nl} - V(r)] R_{nl}(r) = 0$$

where r stands for the relative radial coordinates. The radial wave function $R_{nl}(r)$ is well behaved at the boundaries (the finiteness of the solution requires that $R_{nl}(0) = R_{nl}(r \rightarrow \infty) = 0$) and the transformation

$$u_{nl}(r) = r * R_{nl}(r) \quad (2.14)$$

Reducing radial equation to the simple form

$$\left[-\frac{\hbar^2}{2m_p^*} \frac{d^2}{dr^2} + \frac{\hbar^2 l(l+1)}{2m_p^* r^2} + \frac{V_0}{2R_p^2} r^2 - E_{nl}(r) \right] u_{nl}(r) = 0 \quad (2.15)$$

The general solution of this radial equation is

$$R_{nl}(r) = N_{nl} r^l \exp\left(-\frac{\eta r^2}{2}\right) L_{n-l/2}^{l+0.5}(\eta r^2) \quad (2.16)$$

where $\eta^2 = \frac{2m_p^* V_0}{2R_p^2 \hbar^2}$ and $L_b^a(\eta r^2)$ is the generalized Laguerre polynomial.

Here, the normalization constant is defined as

$$N_{nl} = \left[\frac{2^{n+l+2} \eta^{l+1.5}}{\sqrt{\pi}} \right] \sqrt{\frac{(\frac{n-l}{2})! (\frac{n+l}{2})!}{(n+l+1)!}} \quad (2.17)$$

where $l = n, n-2, \dots, l_{\min}$ and $l_{\min} = 1$ if l is odd and $= 0$ if l is even.

The expectation value is evaluated by determining the general closed form solution for such states, in terms of Laguerre polynomials, together with corresponding eigen values as

$$\left\langle \varphi_{nlm} \left| \frac{1}{r_i} \right| \varphi_{nlm} \right\rangle = \frac{N_{nlm}^2}{2\eta^{l+1}} \int_0^\infty e^{-x_i} x_i^l \left[L_{\frac{n-l}{2}}^{l+0.5}(x)^2 \right] dx_i \quad (2.18)$$

where $x_i = \eta r_i^2$

$$= \frac{N_{nlm}^2}{2\eta^{l+1}} \sum_k \sum_s \left[\frac{-1^{k+s} \Gamma(k+s+l+1) \Gamma(\frac{n+l+3}{2})^2}{\Gamma(\frac{n-l}{2} - k + 1) \Gamma(\frac{n-l}{2} - s + 1) \Gamma(l+s+1.5) \Gamma(l+j+1.5) k! s!} \right] \quad (2.19)$$

The above model can be extended to quantum mechanical two interacting electrons confined in three-dimensional dot geometry. Due to the fact that electron-electron interactions which are known to be quite important in such quasi-zero-dimensional structures are enhanced

by the presence of an additional confinement arising from the magnetic field. However, the complicated nature of the recursion relations appeared in solving the complex integral is given below

$$\left\langle \varphi_{nlm} \left| \frac{1}{r_{ij}} \right| \varphi_{nlm} \right\rangle = \sum_{i \neq j} \sum_l \sum_m \int_0^\infty |R(r_i)|_{nl}^2 r_i^2 dr \int_0^\infty |R(r_i)|_{n_1 l_1}^2 r_j^2 dr \int |\Upsilon_l^m(\Theta_1 \Phi_1)|^2 d\Omega_1 \int |\Upsilon_l^m(\Theta_2 \Phi_2)|^2 d\Omega_2 \Upsilon_l^{*m}(\Theta_1 \Phi_1) \Upsilon_l^m(\Theta_1 \Phi_1) \quad (2.20)$$

2.2 Relativistic Corrections

Although the relativistic effects in the hydrogen atom due to the motion of electrons is small, still, even small numbers become significant for zero-dimensional quantum dots. The Relativistic corrections are applied to Hamiltonian as first order perturbations and the energy eigen values can be further improved by mass correction, Spin orbit coupling and Darwin term.

2.2.1 Energy Shift due to Spin-Orbit Coupling

This term arises due to the fact that spin of the electron modifies the energy level. Due to spin, electron behaves like a little electromagnet, so a moving magnet interacts with the electric field of nucleus causing a change in the energy states. The Spin orbit interaction can remove the spin degeneracy when the potential for electrons in semiconductors is inversion asymmetric. It can split up spin degeneracy associated with quantum number j even without magnetic field. This term is important to study for semiconducting structures as it enables spin orientation and optical detection. The influence of the electron spin on the charge transport in semiconductor nanostructures has attracted considerable interest in recent years.

We are performing a change of basis to $|lsm_l m_s\rangle$ basis and taking the coupled wave functions as the linear combination of the spin wave functions,

$$\Psi_{nl_j m_j} = \sum_{m_l m_s} \langle lsm_l m_s | jm_j \rangle \Psi_{nl m_s} \quad (2.21)$$

where $\langle lsm_l m_s | jm_j \rangle$ are Clebsch-Gordan coefficients. The first order energy corrections (ΔE_{so}) are computed as

$$\Delta E_{so} = \langle \Psi_{nl_j m_j} | H_{SO} | \Psi_{nl_j m_j} \rangle \quad (2.22)$$

$$\Delta E_{so} = \left\langle \Psi_{nl_j m_j} \left| \sum_{i=1}^N \frac{1}{2m_p^* c^2 r_i} \frac{dV_c}{dr} \vec{L} \cdot \vec{S} \right| \Phi_{nl_j m_j} \right\rangle \quad (2.23)$$

$$V(c) = -\frac{e^2}{\epsilon_P r}$$

$$\Delta E_{so} = \frac{e^2 \hbar^2}{4\epsilon_P m_p^* c^2} \left\{ j(j+1) - l(l+1) - \frac{3}{4} \right\} \left\langle \Phi_{nl_j m_j} \left| \sum \frac{1}{r_i^3} \right| \Psi_{nl_j m_j} \right\rangle \quad (2.24)$$

2.2.2 Energy Shift due to Relativistic Correction to Kinetic Energy

This term arises due to the modification in the kinetic energy of particle due to the relativistic mass variation with velocity of particle and is given as

$$H_k = -\frac{p^4}{8m_p^* c^2} \quad (2.25)$$

The relativistic correction for the Impurity energy levels is calculated by applying first order perturbation as

$$\begin{aligned} \Delta E_k = \langle \Psi_{nlm_l m_s} | H_K | \Psi_{nlm_l m_s} \rangle = & -\frac{1}{2m_P^* c^2} E_{nl}^2 + \sum_{i=1}^N \gamma^4 \langle \gamma r_i^4 \rangle_{nl} + \sum_{i=1}^N \frac{2e^2 \gamma^2}{\epsilon_P} \langle r_i^1 \rangle_{nl} + \sum \frac{e^4}{\epsilon_P^2} \left\langle \frac{1}{r_i^4} \right\rangle_{nl} \\ & - \sum \frac{2e^2}{\epsilon_P} \langle r_i \rangle_{nl} E_n - \sum 2\gamma^2 E_n \langle r_i^2 \rangle_{nl} \end{aligned} \quad (2.26)$$

2.2.3 Energy Correction due to Darwin Term

This relativistic correction arises from “Zitterbewegung” of electron – giggling – which smears effective potential felt by electron. Darwin term acts only at the origin as for $l \neq 0$ wave function of impurity vanishes at $r = 0$, so we only consider $l = 0$.

$$H_D = \frac{\pi e^2 \hbar^2}{2m_P^{*2} c^2 \epsilon_P} \delta(r_i) \quad (2.27)$$

Here $\delta(r_i)$ is the Dirac delta function and the energy shift due to this term is written as

$$\Delta E_D = \frac{\pi e^2 \hbar^2}{2m_P^{*2} c^2 \epsilon_P} \langle \Psi_{nlm} | \delta(r_i) | \Psi_{nlm} \rangle \quad (2.28)$$

2.3 Thermal Properties of Spherical Quantum Dot

In this section, we have considered the canonical formalism in which the system is allowed to exchange heat from the surroundings at constant Temperature, Volume and the number of particles. A partition function Z is the essential link between the coordinates of microscopic systems and the thermodynamic properties. Once Partition function Z is evaluated appropriately for the two electron system, then all the thermodynamic quantities, such as Helmholtz free energy (F_H), Internal energy (U), Entropy S , etc, are estimates as the different derivatives of Z .

$$Z = \sum_{n=0}^{\infty} \sum_{l=0}^{\infty} e^{-\beta E'_T} \quad (2.29)$$

Here, Temperature $\beta = \frac{1}{K_B T}$, T = Temperature, K_B = Boltzmann constant.

$$E'_T \text{ (Total Energy)} = E_T + \Delta E_D + \Delta E_K + \Delta E_{SO} \quad (2.30)$$

After obtaining the Partition function Z appropriately for the two electron system, we can estimate all the thermodynamic quantities, such as Helmholtz free energy (F_H), Internal energy (U), Entropy S , etc., as the different derivatives of Z .

$$\text{Internal Energy:} \quad U = -\frac{\log Z}{\partial \beta} \quad (2.31)$$

$$\text{Helmholtz Free Energy:} \quad F_H = -\frac{\log Z}{\beta} \quad (2.32)$$

$$\text{Entropy:} \quad S = \frac{U - F_H}{T} \quad (2.33)$$

The numerical values of the pressure and temperature will be changed to study their effects on the corrected energy values and the thermodynamic potentials in the presence of magnetic field. The pressure- and temperature-dependence is also included in the mass and quantum dot dimensions as

$$R(P) = R_0(1 - (1.5082 * 10^{-3} P)) \quad (2.34)$$

$$m(P) = m_0^* e^{0.078P} \quad (2.35)$$

3. Results

We have studied behaviour of these properties with different confinement strength, Temperature, Pressure and dot radius.

Here, thermodynamic properties such as Internal Energy, Entropy and free energy of a spherical quantum dot has been studied as a function of pressure and dot radius, using effective atomic units (a.u.) for calculations. All the figures have been plotted using both with and without Relativistic effect. Our results are summarized in Figures 1–8. For GaAs, material parameters that have been used are $m(0) = 1$ and $\epsilon(0) = 12.9$ and all the graphs have been plotted for 3 different potentials such are: $V01 = 136.234$ MeV, $V02 = 244.110$ MeV and $V03 = 379.863$ MeV. The main purpose to show both the relativistic and non-relativistic effect is that during initial small values of dot radius, relativistic effect has a negative correction in all the three thermodynamic quantities but for higher values for dot radius the effect is positive and increasing. But in the case of pressure, all the three thermodynamic quantities has an increasing effect from the initial small values and this effect goes on increasing with higher values also.

In Figure 1, the variation of Entropy versus Dot radius (R) for three different confinement potentials at $T = 300$ K has been shown. At smaller effective QD radius, the entropy of the two electrons bounds in the QD increases rapidly with increase in radius. As we increase V_0 , the entropy curve of the two electron system shifts downwards, and it can be attributed to stronger confinement through V_0 . Further as the radius of the QD is increased to almost 30 nm, the changes in the entropy with increase in effective radius becomes less prominent. As the interaction of the electrons is repulsive in nature and is inverse in relationship with relative position, the changes in total internal energy of the electrons in QD are less affected by increase in radius of QD. This therefore manifests in the peculiar shape of the entropy curves.

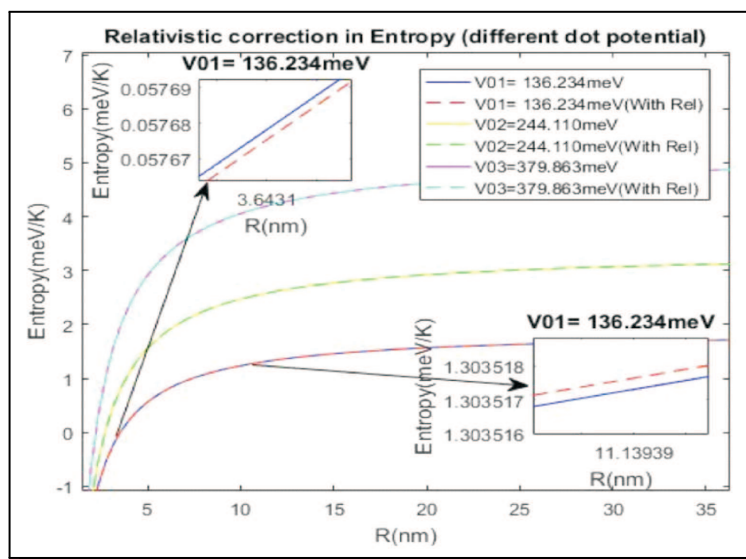


Figure 1

In Figure 2 and 3, we have plotted Entropy vs Pressure for a constant Temperature of 200 K. The main purpose to show the effect of pressure on the entropy was to find the effect on effective

mass and radius of QD. The interesting interplay of effect of pressure on the effective mass and effective radius of the QD results in the near cusp shape of the entropy curves. This interplay is downplayed by the enhancement of V_0 which is shown by the near linear nature of the entropy curve for $V_0 = 244$ as shown clearly in Figure 3 and with enhancement in V_0 the dip moves towards the left of axis.

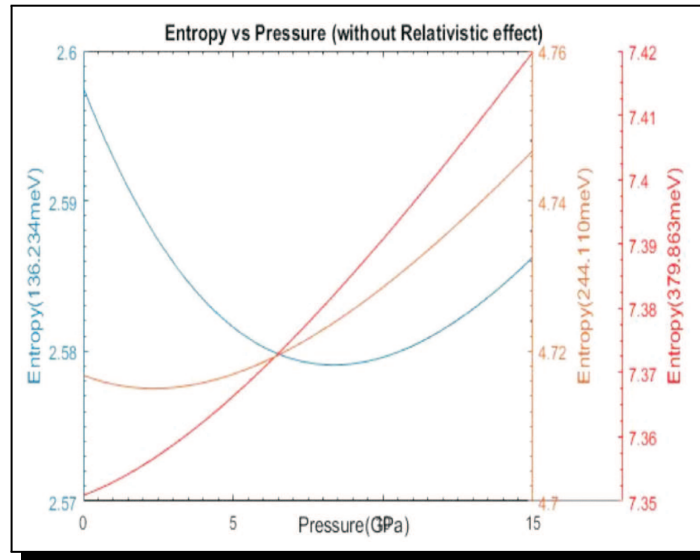


Figure 2

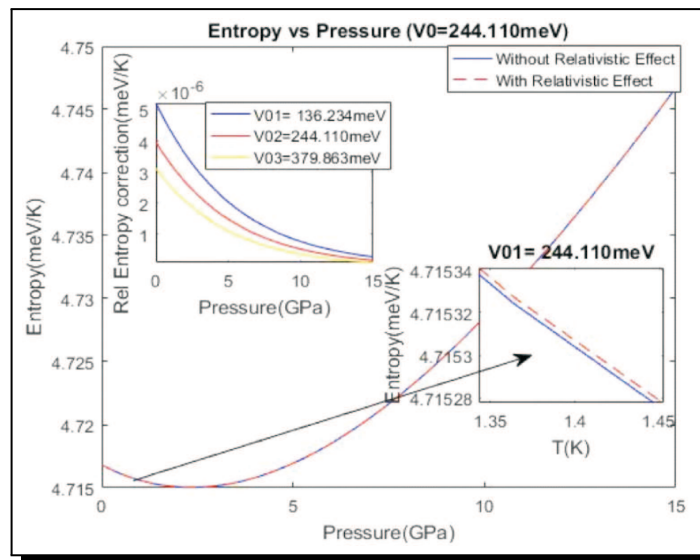


Figure 3

In Figure 4, A plot of Internal Energy versus Dot radius at $T = 300$ K has been shown. Here we see that initially Internal Energy is negative in value because as we know that there is an electron-electron interaction at lower energy levels and at higher levels with increase in QD radius the kinetic energy value is more dominant than the coulomb potential energy, hence

internal energy increasing with increase in effective QD radius. Here the effective radius is less prominent at the higher values than in the beginning. Also it can be observed that the difference in relativistic and non-relativistic energies is of the order of approx. 10^{-5} nm as seen in the closet.

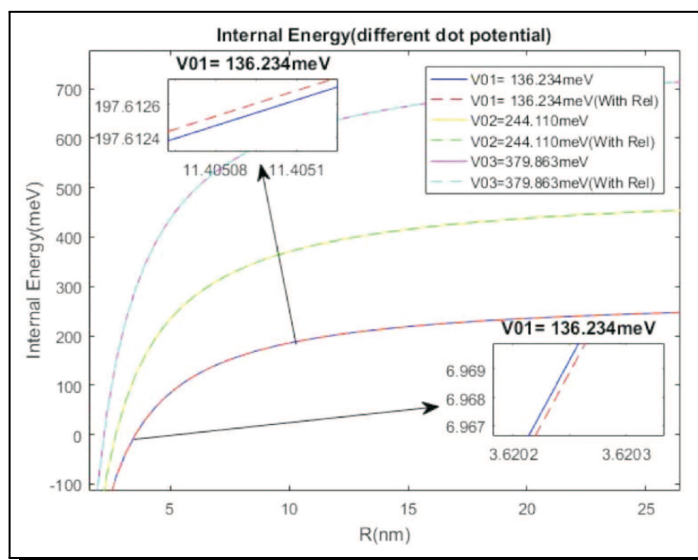


Figure 4

In Figure 5 and 6, variation of internal energy versus pressure for the different potentials for a fixed temperature of 300 K has been shown. The effect of pressure on the effective mass and effective radius of QD results in the dip in the internal energy curve. This effect is more prominent at the lower potential, as with increase in potential the dip vanishes completely and shift towards the left axis, which can be seen in Figure 6.

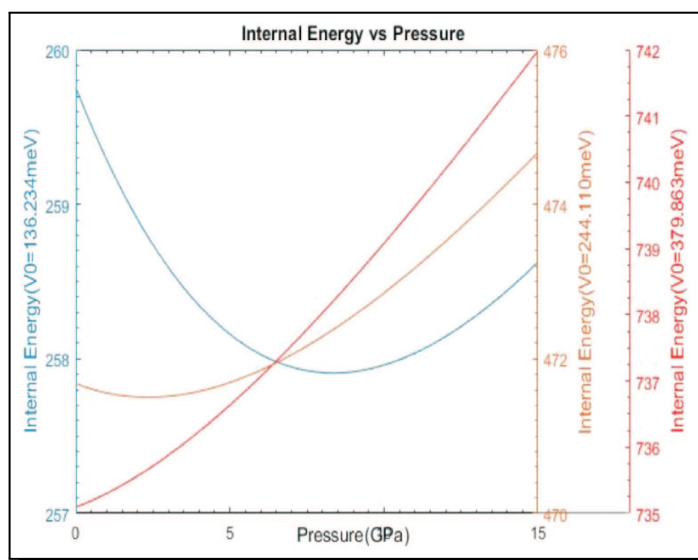


Figure 5

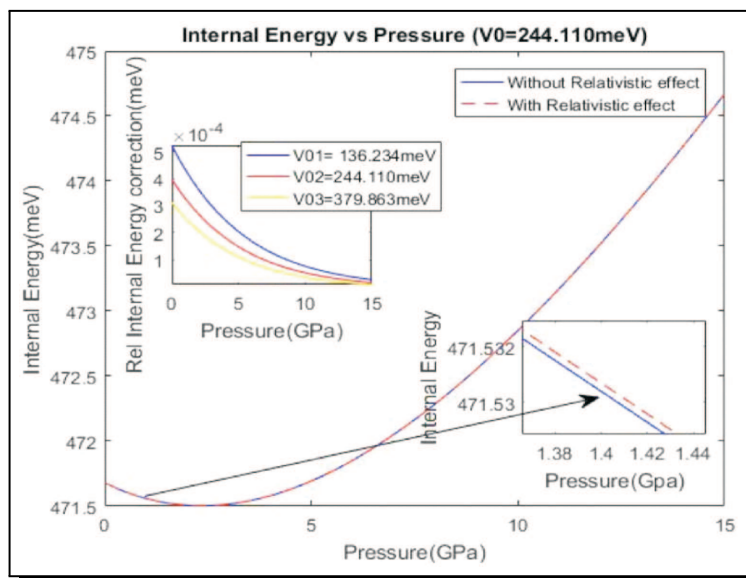


Figure 6

In Figure 7 and 8, plot of free energy vs pressure at a constant temperature $T = 200$ K has been shown. Valuable information which free energy gives is how likely the system transformation from one state to another state spontaneously takes place. Figure 7 shows the pressure dependence of the free energy for the QD. As it can be predicted from both the graphs that free energy is negative for pressure and also increasing with increase in pressure. It is common to have a decreasing trend of free energy and during the progression of any natural process it becomes more negative. The degree of decrease in free energy is estimated by the entropy (S) of the system, where the slope of free energy curve is a negative entropy.

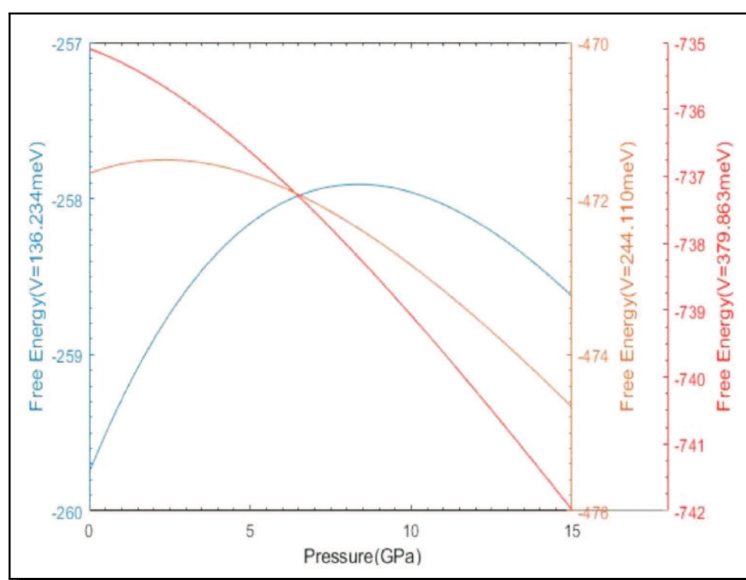


Figure 7

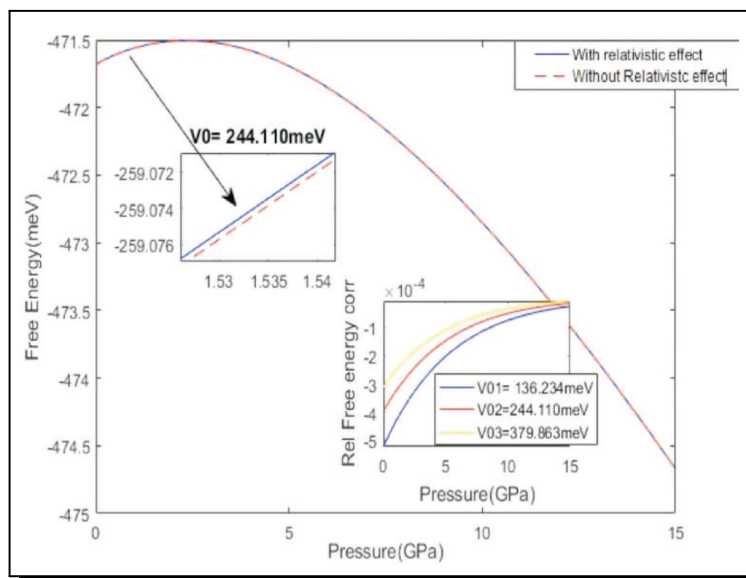


Figure 8

4. Discussion

In conclusion, we have calculated the discrete energy spectra for two electrons in a two-dimensional harmonic oscillator that serves as a simple but suitable model for quantum dots on semiconductor interfaces. We have investigated analytically the thermodynamic properties of a quantum dot as a function of radius without any effect of external magnetic and electric fields. By imposing the relativistic corrections of Pauli's *spin-orbit* (SO) coupling, Darwin's correction, and *mass-velocity* (MV) interaction, we computationally study the electronic structure of an electron confined in a spherical *quantum dot* (QD), and discuss the influence of these relativistic terms. The results have been presented at near room temperatures. We have found that the internal energy and entropy is increasing with the QD radius but there is a slight dip in both the entropy and internal energy when calculated with pressure which vanishes with increase in the potential. The relation of Helmholtz free energy with pressure is negative in nature with the increase in values and is having the slope of the curve negative to that of entropy.

Acknowledgment

Suman Dahiya is thankful to Delhi Technological University, Delhi, India for the financial support. The authors would also like to express their heartfelt gratitude to Prof. Manmohan and Dr. Arun Goyal, University of Delhi for their encouragement and fruitful discussions towards the successful completion of the work.

Competing Interests

The authors declare that they have no competing interests.

Authors' Contributions

All the authors contributed significantly in writing this article. The authors read and approved the final manuscript.

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