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Off-plane Impurity Effects in Graphene Quantum Dots

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Abstract. Within the continuum model of graphene, we study the properties of energy spectrum of graphene quantum dots in the presence of an off-plane donor/acceptor Coulomb impurities as a function of their positions. We propose a variational scheme in which the trial wave functions take into account the confinement of the carriers of graphene quantum dot together with the influence of magnetic field. The dependence of graphene quantum dot states on the location of impurity is investigated in the presence of magnetic field. We show that the off-plane donor/acceptor hydrogenic impurity removes the degeneracy of the relativistic Fock-Darwin states, and modifies the valley splitting due to the spatial confinement.

Keywords: Graphene quantum dots, Hydrogenic impurity

Grafen Kuantum Noktalarında Düzlem Dışı Safsızlık Etkileri

Özet. Grafenin sürekli modeli çerçevesinde, grafen kuantum noktalarının enerji spektrumu konumlarının bir fonksiyonu olarak düzlem dışı donör/akseptor Coulomb safsızlıklarının varlığında çalışılmıştır. Manyetik alanın etkisi altındaki grafen kuantum noktasının yük taşıyıcılarının sınırlanması göz önüne alınarak, varyasyonel yöntemde kullanılacak dalga fonksiyonu önerilmiştir. Grafen kuantum nokta durumlarının safsızlığın konumuna bağlılığı manyetik alanın varlığında incelenmiştir. Düzlem dışı donör/akseptör hidrojenik safsızlığın, göreli Fock-Darwin durumlarının dejenereliğini kaldırdığı ve uzaysal sınırlamadan dolayı vadi dejenereliğinin değiştiği görülmüştür.

Anahtar Kelimeler: Grafen kuantum noktaları, Hidrojenik safsızlık

I. INTRODUCTION

After the discovery of graphene [1-2], there has been great interest in studying the effects of impurities (adatoms) in graphene based nanostructures. Due to the special properties of these structures various impurity models have been presented to understand their role in graphene. Most of theoretical studies [3-19] considers either the continuum model [20] or the tight-binding scheme [21]. It is well-established that, by experiments [13] with adatoms in monolayer graphene, long range impurities are responsible for limiting the mobility of graphene [15], thus Coulomb like hydrogenic impurities are in the center of associated literature [7-10,15]. Since adatoms play important roles on various electronic and optical properties of graphene, studying their role in graphene quantum dots (GODs) would provide better physical insight to the electronic properties of these structures [22-65].

The problem of the Dirac electrons bound to a hydrogenic impurity located at the center of graphene dot (QD) is an interesting problem itself from the point of view of fundamental physics in the pristine graphene. It is originally considered by Pereira *et al* [5]. They have studied the problem of an unscreened Coulomb charge in graphene by solving the problem in continuum description in terms of the 2D Dirac equation. Afterwards, a number of theoretical and experimental works in the graphene literature have been devoted to study of impurity states in pristine graphene. It is expected that, upon the reduction of the size of graphene in making GQDs, impurity related effects become more pronounced.

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In the present paper, we calculate the low-lying energy spectra of the Dirac electron with an off-plane impurity under the influence of a static magnetic field. We introduce a variational scheme based on a choice of trial wave function from the exactly solvable part of the Hamiltonian, and then calculate the energy spectra. It is found that the scalar parabolic confinement together with an off-plane impurity have significant effects on the graphene spectrum. It is also shown that the results obtained within the approximation presented here reduce to the well-known relativistic Landau Levels (LLs) in the absence of both impurity and spatial confinement.

II. THEORY

In the low-energy approximation, the Hamiltonian of a single off-plane hydrogenic impurity in GQD with parabolic confinement under the homogenous magnetic field along the z-axis in graphene quantum dot (GQD) is written as

$$\mathcal{H} = v_F\, \boldsymbol{\alpha}\cdot\left(\boldsymbol{p}+\frac{e}{c}\,\boldsymbol{A}\right)+\boldsymbol{\beta}\Delta_0 r^2-\frac{Ze^2}{\epsilon\sqrt{r^2+z_0^2}} \tag{1}$$

where $\Delta_0 = U_0/2R_0^2$. In Eq.(1), $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}$ are the Dirac matrices while U_0 and R_0 are the confining strength and the radius of the GQD respectively, and $v_F = (3a/2)J_0$ and z_0 are the Fermi velocity and the position of impurity from the dot center chosen to be $r_i = (0,0,z_0)$. Here, J_0 is the resonance integral between nearest neighbor carbon atoms, and is of order of 2.7 eV, and a is the equilibrium bond length of the graphene. $\boldsymbol{A}$ is the vector potential in the symmetric gauge, i.e., $\boldsymbol{A} = B_0(-\mathrm{y},\mathrm{x},0)/2$ corresponding to a magnetic field perpendicular to GQD plane. The solution of Eq.(1) in the absence of GQD potential is well established Ref.17. In this case, the eigenfunctions corresponding to the Dirac-Weyl equation $\mathcal{H}\Psi_\mathrm{n} = \mathrm{E_n}\Psi_\mathrm{n}$ which are four-component spinors are given by

$$\begin{aligned}\Psi_n^{\boldsymbol{K}}(x,\varphi) &= R_n(x)e^{im\varphi}e^{-x^2/2}\begin{pmatrix}\mathbb{L}^0\\0\\0\\\mathbb{L}^{\boldsymbol{K}}\end{pmatrix}\\ \Psi_n^{\boldsymbol{K'}}(x,\varphi) &= R_n(x)e^{im\varphi}e^{-x^2/2}\begin{pmatrix}0\\\mathbb{L}^0\\\mathbb{L}^{\boldsymbol{K'}}\\0\end{pmatrix}\end{aligned} \tag{2}$$

with $R_n(x) = [\nu!/2\pi(\nu+|m|)!]^{1/2}\gamma_0 x^{|m|}$. In Eq. (2) we have defined $\mathbb{L}^{\boldsymbol{0}}$, $\mathbb{L}^{\boldsymbol{K}}$ and $\mathbb{L}^{\boldsymbol{K'}}$ in terms of Laquerre polynomials as $\mathbb{L}^{\boldsymbol{0}}(x) = \mathbb{L}_\nu^{|m|}(x^2)$,

$$\begin{aligned}\mathbb{L}^{\boldsymbol{K}} &= \frac{ixe^{+i\varphi}}{\sqrt{n}}\begin{cases}L_\nu^{|m|+1}(x^2) & m\geq 0\\ -\dfrac{n}{x^2}L_{\nu+1}^{|m|-1}(x^2) & m<0\end{cases}\\ \mathbb{L}^{\boldsymbol{K'}} &= \frac{ixe^{-i\varphi}}{\sqrt{n}}\begin{cases}-\dfrac{n}{x^2}L_\nu^{|m|-1}(x^2) & m\geq 0\\ L_{\nu+1}^{|m|+1}(x^2) & m<0\end{cases}\end{aligned} \tag{3}$$

respectively. The corresponding eigenvalues are given by $E_n = \lambda\epsilon_n$ wherein $\epsilon_n = \hbar v_F\sqrt{2n}/l_B$, λ is the chirality index and takes -1 and $+1$ values, which correspond to valence and conduction bands of graphene, respectively. $l_B = (\hbar c/eB)^{1/2}$ is the magnetic confinement length, and $n = \nu + (|m| + m + \tau + 1)/2$ where τ is considered as a pseudospin index whose -1 and $+1$ values specify the two valleys, i.e., $\boldsymbol{K}$ and $\boldsymbol{K}'$, respectively. In the following, we will restrict ourselves to only conduction band. Extension of the results to the other band is straightforward. In Eqs.(2) and (3), $x = \gamma_0\rho$ is a dimensionless, and γ_0 is the inverse of characteristic length of the system. On the one hand, without spatial parabolic confinement, γ_0 is proportional to inverse of magnetic confinement length l_B, i.e., $\gamma_0 = \gamma = 1/\sqrt{2}l_B$. On the other hand, in the presence of spatial confinement, graphene system described by the Hamiltonian Eq.(1) suggests a natural unit of length $R = (\hbar v_F R_0^2/U_0)^{1/3}$ which characterizes the size of a scalar parabolic quantum dot in graphene [22-23] . We will use the ansatz of Ref. 23 to characterize the system by only one parameter, i.e., we write $\gamma_0^2 = \gamma^2 + R^{-2}$ and take γ to be a variational parameter, so as to reduce to $1/\sqrt{2}l_B$ in the limit $R \to \infty$. Therefore, the variational eigenenergies of GQD is given by

$$E_n = \int d^2\boldsymbol{r}\, \Psi_n^\dagger \left[\boldsymbol{\alpha}\cdot\left(-i\boldsymbol{\nabla} + \frac{e}{\hbar c}\boldsymbol{A}\right) + \boldsymbol{\beta}\Delta_0 r^2 - \frac{Ze^2}{\epsilon\sqrt{r^2+z_0^2}}\right]\Psi_n \tag{4}$$

If the Eq.(2) is replaced into Eq.(4) by using the relation

$$\frac{1}{\sqrt{r^2+z_0^2}} = \frac{2}{\sqrt{\pi}}\int_0^\infty e^{-(r^2+z_0^2)u^2}du$$

after some derivations, the variational energy can be written as

$$\tilde{E} = \left(\sqrt{n} - \frac{1}{2}\bar{Z}\widetilde{M}_n^I\right)\gamma_0 + \frac{\sqrt{n}}{2\bar{l}_B^2}\frac{1}{\bar{\gamma}_0} + \frac{1}{4\bar{R}^3}\frac{1}{\bar{\gamma}_0^2}\widetilde{M}_n^{II} \tag{5}$$

where we have defined the integral $\widetilde{M}_n^I$ in Eq.(5) as

$$\widetilde{M}_n^I = \frac{2}{\sqrt{\pi}}\frac{\nu!}{(\nu+|m|)!}\int_0^\infty e^{-\bar{z}_0^2\bar{\gamma}_0^2u^2}du\left[\int_0^\infty dr\; r^{|m|}e^{-(1+u^2)r}\left|L_\nu^{|m|}(r)\right|^2 + \frac{1}{n}\int_0^\infty dr\; r^{|m|}e^{-(1+u^2)r}R_\nu^{K(K\prime)}(r)\right]$$

with

$$R_n^K = \begin{cases} r\left|L_\nu^{|m|+1}(r)\right|^2 & m \geq 0 \\ n^2 r^{-1}\left|L_{\nu+1}^{|m|+1}(r)\right|^2 & m < 0 \end{cases}$$

$$R_n^{K\prime} = \begin{cases} n^2 r^{-1}\left|L_\nu^{|m|-1}(r)\right|^2 & m \geq 0 \\ r\left|L_{\nu-1}^{|m|+1}(r)\right|^2 & m < 0 \end{cases}$$

$\widetilde{M}_n^{II}$ in Eq.(5) arising from the GQD potential includes integrals in terms of associated Laguerre polynomials and it can be written in closed form as

$$\widetilde{M}_n^{II} = \mathbb{F}_n - \frac{1}{n}\mathbb{G}_n$$

where

$$\mathbb{F}_n = \frac{\nu!}{(\nu+|m|)!}\int_0^\infty dr\ r^{|m|+1}\, e^{-r}\, L_\nu^{|m|}(r^2)$$

$$\mathbb{G}_n = \frac{\nu!}{(\nu+|m|)!}\int_0^\infty dr\ r^{|m|+1}\, e^{-r}\, \mathbb{H}_{\nu,\alpha}^{|m|}(r)$$

with

$$\mathbb{H}_{\nu,\alpha}^{|m|}(r) = \begin{cases} r\left|L_{\nu+\alpha}^{|m|+1}(r)\right|^2, & \alpha = \begin{cases} 0 & m \geq 0\ \ \tau = +1 \\ -1 & m < 0\ \ \tau = -1 \end{cases} \\ \dfrac{n^2}{r}\left|L_{\nu+\alpha}^{|m|-1}(r)\right|^2, & \alpha = \begin{cases} 0 & m \geq 0\ \ \tau = +1 \\ +1 & m < 0\ \ \tau = -1 \end{cases} \end{cases}$$

where $L_\nu^{|m|}(r)$ are the associated Laguerre polynomials. Integrals in $\widetilde{M}_n^{II}$ can be integrated separately [66] to yield $\mathbb{F}_n = 2\nu + |m| + 1$ and $\mathbb{G}_n = 2n - m$, respectively. Thus, it reduces to $\widetilde{M}_n^{II} = -\tau$. Before numerical minimization of Eq.(5), to get an approximate analytical expression for the energy we investigate some its limiting cases. Since the minimization of the energy given by Eq.(5), in the absence of both spatial confinement potential and the impurity, with respect to the variational parameter $\bar{\gamma}$ gives $\bar{\gamma} = 1/\sqrt{2}l_B$, by replacing this value back into the relevant variational energy, one gets $\bar{E}_n = \sqrt{2n}/\bar{l}_B$, which are the well-known Landau levels for massless graphene. $\widetilde{M}_n^I$ can also be integrated analytically for each n. For zero energy states, it can be found in terms of confluent hypergeometric functions as $\widetilde{M}_n^I = U\left(\frac{1}{2}, \frac{1}{2} - l, \frac{\bar{z}_0^2}{\bar{l}_0^2}\right)$, where l takes values 0 and $|m|$, respectively. It also reduces to $\Gamma(|m| + 1/2)/\Gamma(|m| + 1)$ when $\bar{z}_0 = 0$. Consequently, substituting these values back into Eq.(5) as a first approximation yields

$$\bar{E}_0 = \frac{-\bar{Z}}{2\bar{l}_0} U\left(\frac{1}{2}, \frac{1}{2} - l, \frac{\bar{z}_0^2}{\bar{l}_0^2}\right) - \tau \frac{\bar{l}_0^2}{4\bar{R}^3} \tag{6}$$

where $1/\bar{l}_0^2 = 1/\bar{l}_B^2 + 1/\bar{R}^2$ defines the effective confinement length. Eq.(6) may give an idea about the effect of off-plane impurity on the GQD spectrum. From Eqs.(6), in the absence of magnetic field for $m = 0$, we estimate the energy

$$\bar{E}_0 = -\frac{1}{2}\bar{Z} e^{\bar{z}_0^2/\bar{R}^2} \Gamma(1/2\,, \bar{z}_0^2/\bar{R}^2) + \frac{1}{4\bar{R}} \tag{7}$$

so that the critical value of $\bar{Z}$ can be easily found that

$$\bar{Z}_{crt} = \frac{1}{2} \frac{e^{\bar{z}_0^2/\bar{R}^2}}{\Gamma\left(1/2, \bar{z}_0^2/\bar{R}^2\right)}. \tag{8}$$

This reduces to $\bar{Z}_{crt} = 1/2\sqrt{\pi}$ at $\bar{z}_0 = 0$. In Eq.(8), we have used the relation $U\left(\frac{1}{2}, \frac{1}{2} - l, \frac{\bar{z}_0^2}{\bar{l}_0^2}\right) = e^{\bar{z}_0^2/\bar{R}^2} \Gamma(1/2\,, \bar{z}_0^2/\bar{R}^2)$. In the absence of impurity, Eq.(7) is consistent with the reported experimental values for the graphene devices fabricated by Ponomarenko et al [67] , wherein the GQDs have a gap value of $0.010 - 0.5\, eV$ for the GQDs with diameter of $D = 40 - 1.0\, nm$. Eq.(6) predicts their corresponding values as $0.014 - 0.57\, eV$. The valley splitting of energy levels can easily be seen from the FIG. 1(a) and 1(b) as the impurity approaches onto GQD. It should be noted that the degeneracy of relativistic LLs with negative values of angular momentum quantum number m is lifted by the off-plane impurity, and they can be controlled by the position of the impurity. Another obvious feature of the figure is that, as a result of competition of confinement potential and magnetic field, the donor(acceptor) impurity strongly affects the size of the valley splitting, and lowers (raises) it towards smaller (higher) values, with decreasing $\bar{z}_0$. The splitting become higher and higher as approach to the graphene surface.

The binding energy is defined as the minimum energy required to remove the Dirac electron to infinity where impurity strength is zero, i.e., it is simply the difference between energies with and without impurity. The binding energy as a function of dot radius as well as the confinement strengths is plotted in FIG. 2 and in FIG. 3 for a typical value of $\bar{Z} = 0.15$. As is seen from the FIG. 3, binding energies increases with decreasing confinement length. They further increase by switching the magnetic field on, since the magnetic field immerse the electron towards to the impurity center. This can also be controlled by the position of the impurity.

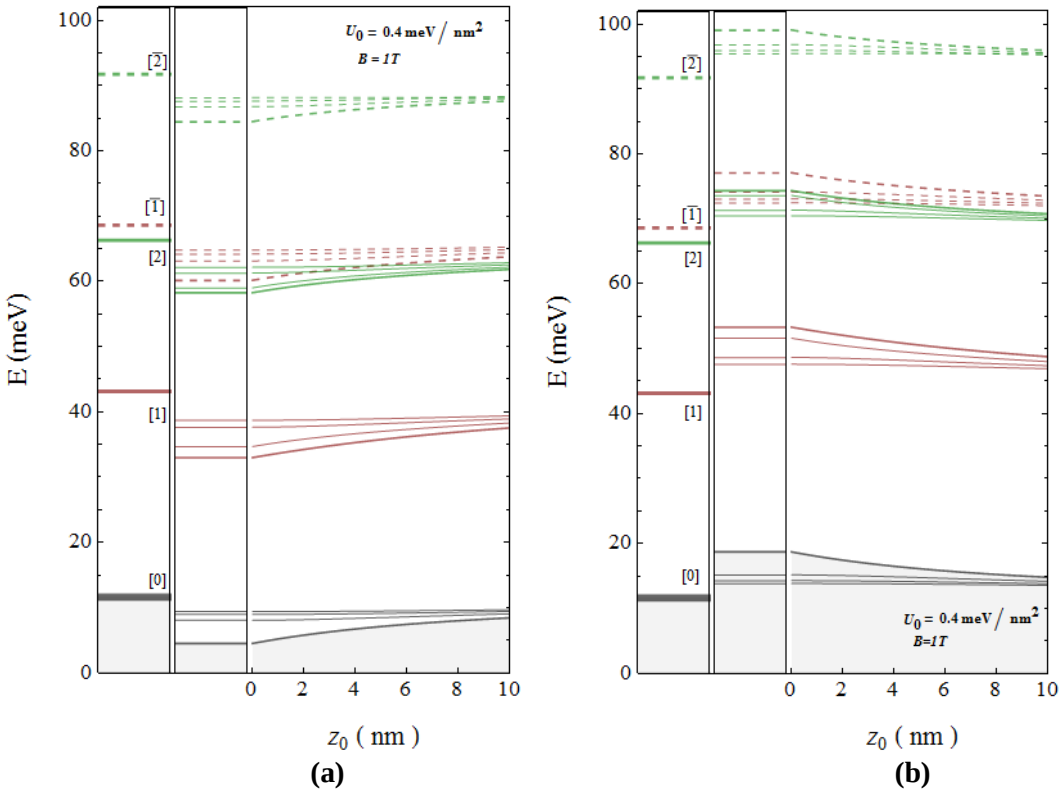

Figure 1. The spectra of low-lying states of GQD electron as a function of position of an **(a)** donor **(b)** acceptor impurity. The numbers in the brackets represent energy levels. The first columns of each of two energy diagrams are for $\bar{Z} = 0$, while second ones are for $\bar{Z} = 0.15$. The solid and dashed curves represent $\boldsymbol{K}$ and $\boldsymbol{K}'$ valleys.

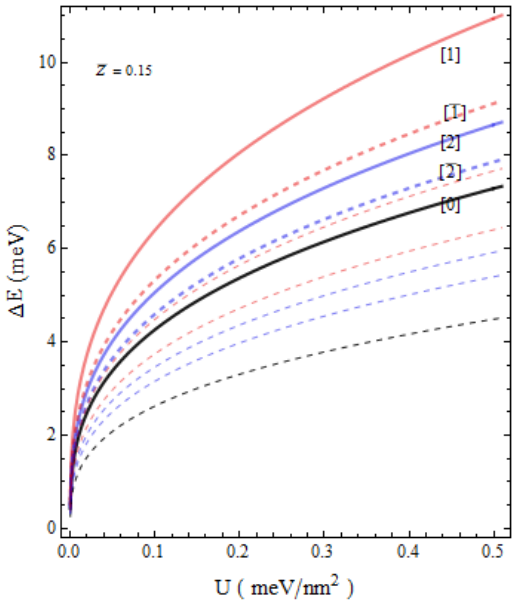

Figure 2. Impurity binding energies as a function of dot strength. While the thick straight (dashed) lines, represented by labels $[n]$ ($[\bar{n}]$), correspond to the $\boldsymbol{K}$ ($\boldsymbol{K}'$) valleys for $z_0 = 0$, the thin straight (dashed) ones correspond to $\boldsymbol{K}$ ($\boldsymbol{K}'$) valleys again, but for $z_0 = R/2$, in the absence of magnetic field.

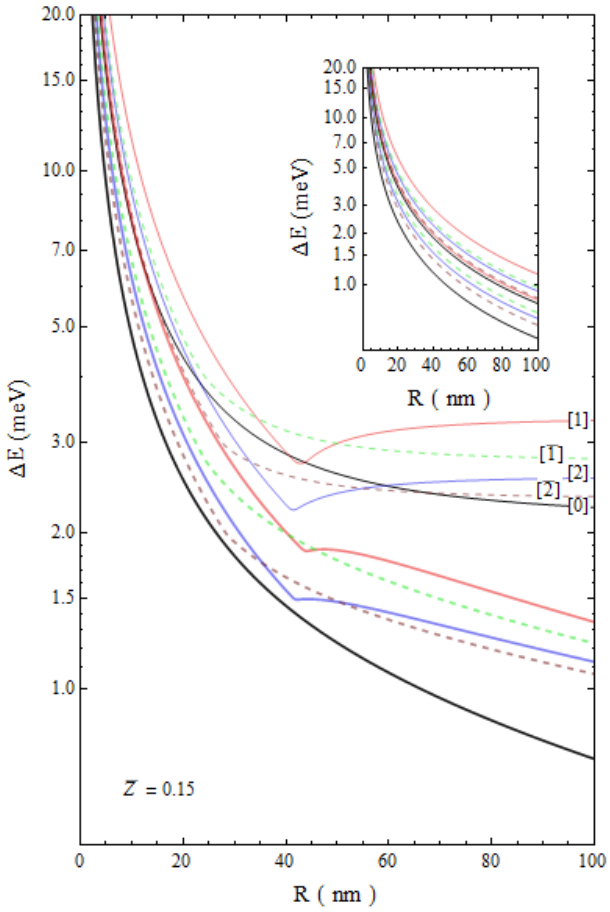

Figure 3. Impurity binding energies associated with the first two GQD levels as a function of GQD radius R in nanometers for $B = 1\,T$ and $\bar{Z} = 0.15$. Curves are for different values of $\bar{Z}$. While the thin straight (dashed) lines, represented by labels $[n]$ ($[\bar{n}]$), correspond to the $\boldsymbol{K}$ ($\boldsymbol{K'}$) valleys for $z_0 = 0$, the thick straight (dashed) ones correspond to the $\boldsymbol{K}$ ($\boldsymbol{K'}$) valleys again, but for $z_0 = R/2$. Inset includes the same graphs in the absence of magnetic field.

III. CONCLUSIONS

In conclusion, we investigated theoretically long range hydrogenic impurity related effects on the electronic energy spectrum of massless Dirac fermions in parabolic QDs. We employ a variational scheme based on using exact solutions of Dirac-Weyl equation in the absence of quantum dot potential as trial wave functions. The electronic energy spectrum of the Dirac electron bound to a hydrogenic off-plane impurity subjected to a uniform magnetic field together with the impurity binding energies are all calculated. The results show that, due to the presence of off-plane impurity, the magnitude of the valley splitting can easily be controlled. It starts to enhance away from the plane, allowing to control the the splitting. In the high magnetic field regime, we also checked that the validity of the obtained results by reproducing the well-known gapless spectrum of the graphene in the absence of impurity.

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