Numerical method for *N* electrons bound to a polar quantum dot with a Coulomb impurity

J. K. F. Yau* and C. M. Lee†

Superconductor Fabrication Laboratory, Department of Manufacturing Engineering and Engineering Management, City University of Hong Kong, Hong Kong, People's Republic of China

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A numerical method is proposed to calculate the Frohlich Hamiltonian containing *N* electrons bound to polar quantum dot with a Coulomb impurity without transformation to the coordination frame of the center of mass and by direct diagonalization. As an example to demonstrate the formalism of this method, the low-lying spectra of three interacting electrons bound to an on-center Coulomb impurity, both for accepter and donor, are calculated and analyzed in a polar quantum dot under a perpendicular magnetic field. Taking polaron effect into account, the physical meaning of the phonon-induced terms, both self-square terms and cross terms of the Hamiltonian are discussed. The calculation can also be applied to systems containing particles with opposite charges, such as excitons.

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I. INTRODUCTION

The theoretical studies of quantum dots containing few electrons and excitons become a growing interest since self-assembled quantum dots can be fabricated in laboratories.[1–15] The great interests are due to the potential use in designing electro-optical devices. The properties of the dots including impurities are one of the interests in this area because they modify the energy levels of the materials and in turn affect their electronic and optical properties.

The theoretical studies of few electrons in two-dimensional space and a perpendicular magnetic field can be traced back to the early 1980's. The three-electron system with parabolic pressure was first investigated by Laughlin in the context of fractional Hall effect.[16] His work became the groundwork of the study of quantum dots. He constructed the spin-polarized correlated states in the lowest Landau levels. Similar works was later carried out up to seven electrons.[17,18] Using the method of numerical diagonalization, the energy spectra of quantum dots up to eight electrons were also calculated.[8–11] The role of the electron correlation, the effects of confinement and external magnetic field were discussed in great detail. Furthermore, energy spectra and persistent currents of few-electron quantum rings were also studied.[19–23] However, the above studies only focused on the Coulomb interaction of electrons. They neither consider the effect of phonon nor the effect of impurities.

Polaron Hamiltonian was first derived by Frohlich[24] using the quantum theory of field on the basis of electron-phonon interaction. This Hamiltonian was then solved by Lee-Low-Pines method[25] via two subsequent unitary transformations: the first transformation is to eliminate the center-of-mass coordinates, while the second is to displace the phonon coordinates. In the last two decades, the polaron Hamiltonians of one and two electrons in two-dimensional systems, such as quantum wells,[26–29] one-dimensional systems, such as quantum wires,[30,31] and zero-dimensional systems, such as quantum dots,[12,32–36] were investigated. Besides these works on one- or two-electron systems or in the studies of exciton, the Hamiltonians were also first transformed to center-of-mass frame before performing the unitary transformations. Taking the impurity into account, the early investigation of impurity-phonon interaction effects on these low-dimensional structures was reviewed in Ref. 37. Within the framework of variation approach, the binding energies of the ground state of a hydrogenic donor as a function of the size of the quantum well were first calculated.[38] The studies of the same framework were then modified[39] and extended to cylindrical,[40] rectangular,[41] infinite and finite cylindrical[42] quantum wires, and even quantum dots.[13,14] Since the existence of on-center D^- in GaAs/Al_xGa_{1-x}As quantum wells was first reported using far-infrared magnetotransmission and magnetophotoconductivity measurements,[43] both experimental and theoretical studies in quantum wells and quantum dots have been carried out intensively.[44,45] However, the theoretical studies of electron-phonon systems with more than two electrons are rare.

The objective of the present study is to develop a numerical method to calculate the low-lying energy spectra of *N*-electron polar quantum dot bound to an on-center Coulomb impurity. This is a case that *N* electrons in a quantum dot interact with longitudinal-optical (LO) phonon and an on-center Coulomb impurity in a magnetic field. The calculation is an example of utilizing this method taking $N=3$. The evolutions of the ground-state energy induced by magnetic fields, magnetic moment, and magnetic susceptibility are reported for the total electrons' spins $S=1/2$ and $S=3/2$, respectively. The modification of the Hamiltonian by the phonon induced terms with the phase factors for the three electrons is discussed. It is worth noting that, in the present study, the method can also be applied to those systems with the particles of opposite charges, e.g., excitons. In the latter case, the difference is that the opposite signs are added for the phase factors of those particles with opposite charges.

II. THEORY

Electrons moving in a quantum dot in the presence of a magnetic field are considered as confined in a two-dimensional (2D) parabolic potential defined as[17] $\frac{1}{2}m\omega_o^2\rho_i^2$, where $\vec{\rho}_i$ is the 2D position vector of the electron *i*. In the case that *N* electrons bound to a Coulomb impurity in the

dot, they interact not only with the surrounding field of LO phonon but also with the Coulomb potential under the magnetic field B along the z direction. Taking the effective-mass approximation, the Hamiltonian for such a system of N electrons is given by

$$\hat{H}=\hat{H}_0+\hat{H}_{\rm co}+\hat{H}_{\rm imp}+\hat{H}_{\rm ind}+\hat{H}_{\rm spin}, \tag{1}$$

in which the unperturbed term $\hat{H}_0$, the Coulomb repulsion term between electrons $\hat{H}_{\rm co}$, the Coulomb interaction term between electron and impurity $\hat{H}_{\rm imp}$, the phonon induced term $\hat{H}_{\rm ind}$, and the spin term are expressed as

$$\hat{H}_0=\sum_{i=1}^{N}\left[\frac{1}{2m}[\vec{P}_i+e\vec{A}(\vec{\rho}_i)]^2+\frac{1}{2}m\omega_o^2\rho_i^2\right], \tag{2}$$

$$\hat{H}_{\rm co}=\sum_{i<j}\frac{e^2}{\epsilon_\infty|\vec{\rho}_i-\vec{\rho}_j|}, \tag{3}$$

$$\hat{H}_{\rm imp}=\sum_{i=1}^{N}\frac{\eta e^2}{\epsilon_\infty\rho_i}, \tag{4}$$

$$\hat{H}_{\rm ind}=\sum_{\vec{q}}\hbar\omega_{LO}\hat{a}_{\vec{q}}^{+}\hat{a}_{\vec{q}}+\sum_{\vec{q}}\{\hat{a}_{\vec{q}}V_{\vec{q}}(\vec{\rho}_1,\vec{\rho}_2,\ldots,\vec{\rho}_N)+\text{H.c.}\}, \tag{5}$$

$$\hat{H}_{\rm spin}=g^*\mu_B BS_z. \tag{6}$$

The meanings of the symbols in Eq. (1) are as follows: As before, $\vec{\rho}_i$ is the 2D position vector of electron i; ω_o is the confining strength of the quantum dot; the parameter η is 1 for an accepter impurity and -1 for a donor impurity; and $\hat{a}_{\vec{q}}^{+}(\hat{a}_{\vec{q}})$ is the creation (destruction) operator of the phonon with 3D wave vector $\vec{q}=(\vec{q}_\parallel,q_z)$ in optical branch. $\hat{H}_{\rm spin}=g^*\mu_B BS_z$ is the Zeeman energy for the coupling of the spins of charged particles and the magnetic field B. μ_B is the Bohr magneton, g^* is the effective Lande factor, and S_z is the z component of the total spins of the N electrons.

We model the interaction between the phonons and the electrons by the corresponding bulk modes[46] in the quantum dot with the neglection of the quantization of phonons due to the size of the dot for mathematical simplicity. In a system of N electrons all with the same mass, the coupling strength with phonon is given by

$$V_{\vec{q}}(\vec{\rho}_1,\vec{\rho}_2,\ldots,\vec{\rho}_N)=\sum_{i=1}^{N}\zeta_i V_{\vec{q}}e^{i\vec{q}\cdot\vec{\rho}_i}, \tag{7}$$

with all the parameters $\zeta_i=1$ and the coefficient of electron-phonon coupling[24] $V_{\vec{q}}$ which depends on the electron (hole)-LO phonon coupling constant α_{LO}. This strength contains N phase factors, and $\zeta_i=-1$ for those corresponding particles with opposite charges.

Using the symmetry gauge description for the magnetic field B, the vector potential $\vec{A}(\vec{\rho}_i)=(-\frac{1}{2}By_i,\frac{1}{2}Bx_i,0)$ is chosen and the unperturbed Hamiltonian can be rewritten as

$$\hat{H}_o=\sum_{i=1}^{N}\left(\frac{P_i^2}{2m}+\frac{1}{2}m\omega_B^2\rho_i^2+\frac{1}{2}\omega_c L_i\right), \tag{8}$$

in which $\omega_B=\sqrt{\omega_o^2+\omega_c^2/4}$, $\omega_c=eB/m$ is the cyclotron frequency, L_i is the angular momentum of the individual electron.

In order to diagonalize the phonon induced term of the Hamiltonian $\hat{H}_{\rm ind}$ at zero temperature, the unitary operator[25] $\hat{U}$ is introduced to displace the phonon coordinates

$$\hat{U}=\exp\left[\sum_{\vec{q}}(f_{\vec{q}}\hat{a}_{\vec{q}}^{+}-f_{\vec{q}}^{*}\hat{a}_{\vec{q}})\right] \tag{9}$$

where the displacement amplitude $f_{\vec{q}}$ and its complex conjugate $f_{\vec{q}}^{*}$ are to be determined by minimizing the matrix elements $\langle 0_{\rm ph}|\hat{U}^{-1}\hat{H}\hat{U}|0_{\rm ph}\rangle$ of the Hamiltonian, and $|0_{\rm ph}\rangle$ is the phonon ground state at absolute zero temperature.

To obtain the eigenfunction and the eigenenergies, for the case of N electrons, we diagonalize $\hat{H}$ in a model space spanned by a set of the translationally invariant 2D harmonic product basis functions with a total orbital angular momentum L $(=l_1+l_2+\ldots+l_N)$, a parity π and a total spin S in the form of a Slate determinant

$$\psi^{L,\pi}=\frac{1}{\sqrt{N!}}\begin{vmatrix}\phi_{n_1l_1}(\vec{\rho}_1)\chi_1 & \phi_{n_1l_1}(\vec{\rho}_2)\chi_1 & \cdots & \phi_{n_1l_1}(\vec{\rho}_N)\chi_1\\ \phi_{n_2l_2}(\vec{\rho}_1)\chi_2 & \phi_{n_2l_2}(\vec{\rho}_2)\chi_2 & \cdots & \phi_{n_2l_2}(\vec{\rho}_N)\chi_2\\ \cdots & \cdots & \cdots & \cdots\\ \phi_{n_Nl_N}(\vec{\rho}_1)\chi_N & \phi_{n_Nl_N}(\vec{\rho}_2)\chi_N & \cdots & \phi_{n_Nl_N}(\vec{\rho}_N)\chi_N\end{vmatrix}. \tag{10}$$

Here, an interchange of two electrons corresponds to an interchange of two columns of the determinant. As a result, the total wave function $\psi^{L,\pi}$ changes sign. $\phi_{n_il_i}$ and χ_i are the spatial and the spin parts of the electron i, respectively. The one-electron eigenstate $\phi_{nl}(\rho)$ can be expressed in associated Laguerre polynomials with the corresponding eigenvalue $2n+|l|+1$ for an electron moving in a homogeous magnetic field and 2D parabolic potential. n and $l\hbar$ are the

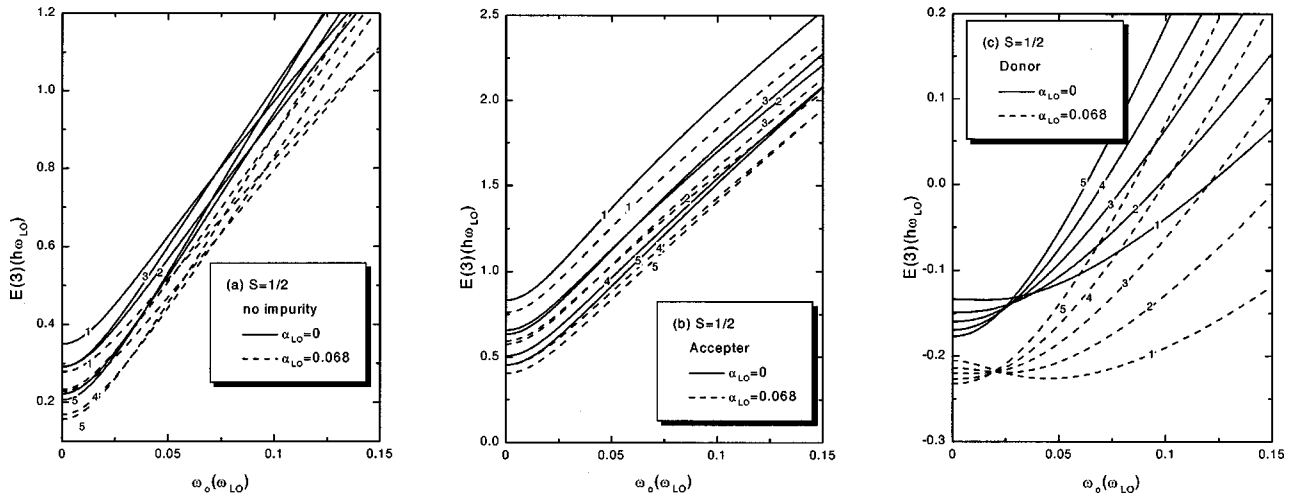

FIG. 1. Lowest-energy states of total orbital angular momentum $L=1,2,3,4,$ and 5 with and without polaron effect as functions of confining strength for the total spin $S=1/2$ under magnetic field $B=1$ T, (a) no impurity, (b) with an accepter impurity, and (c) with a donor impurity.

radial quantum number and the angular momentum, respectively. The symbol $a=\sqrt{\hbar/m\omega_B}$ is the size of the dot.

In our present numerical work (see Appendix), using the number of electrons $N=3$ as an example, the low-lying eigenvalues $E(3)$ of the three-electron quantum dot bound to a Coulomb impurity for $S_z=1/2$ and $S_z=3/2$ are first calculated. Then the magnetic moments and the magnetic susceptibilities of the system are found.

In this work, only the non-negative orbital angular momenta l_i of the individual electrons with zero quantum numbers n_i are considered which do not largely affect the qualitative results.

III. DISCUSSION AND CONCLUSIONS

To what follows, one of the typical semiconducting materials GaAs, is taken as an example to discuss both energy spectra and magnetic moments as a function of magnetic fields. The material parameters are as follows:[47] $\epsilon_o=12.83$, $\epsilon_\infty=10.90$, $\hbar\omega_{LO}=36.7$ meV, Lande factor $g^*=0.44$, $m=0.675m_o$, where m_o is the single-electron bare mass.

In the case that three electrons confined in a quantum dot, the electrons position themselves form a regular triangle. The configuration is known to be energetically stable[9] and the electrons approach each other when the confinement strength ω_o increases. In order to show the ω_o dependency of eigenenergy of such a system, the low-lying states $L=1-5$ for the unpolarized state ($S_z=1/2$) in a fixed magnetic field $B=1$ T as a function of ω_o are plotted with and without impurity, either accepter or donor. Figure 1 shows the results with phonon correction by dashed lines and the results without phonon correction by solid lines. It is obvious that the eigenenergies when phonon effect are taken into account are lower. Hence, the system of three electrons is more stable with the effect of phonon-electron interaction.

The second note to make is that, for the dot with a donor impurity [Fig. 1(c)], at weak confinement strength (when ω_o is below about $0.25\omega_{LO}$), the state $L=1$ has smaller centrifugal barriers such that the three electrons are closer to each other so that the forces between them are more repulsive. As is known, a single-electron eigenstate $|\phi_{nl}\rangle=(1/\sqrt{2\pi})e^{il\theta}R_{nl}(r)$ can be qualitatively regarded as the circular motion of the electron about the center of the quantum dot with orbital angular momentum $l\hbar$. The mean-square orbit radius r can be expressed as,[9]

$$\langle\phi_{nl}|r^2|\phi_{nl}\rangle\propto 2n+|l|+1. \tag{11}$$

The lower the angular momentum state l, the closer the electrons to the center of the quantum dot. From Eq. (11), one can see that the trend of eigenenergy $E(l)$ for electrons with positive angular momenta l is the same as those with negative l. For simplicity, we consider the electrons with non-negative angular momentum l in the present calculation. The state $L=0$ is forbidden according to Pauli exclusion principle although it has no centrifugal barriers. The state $L=1$ is therefore most unstable comparing with the other angular momentum states at lower ω_o. At higher ω_o, the energy spectra are, however, different from those of low ω_o, especially for donor doped quantum dot. In Fig 1(c), it can be seen that there is a clear-cut transition point at $\omega_o\approx 0.035\omega_{LO}$ without polaron effect and $0.025\omega_{LO}$ with polaron effect. The higher the ω_o, the closer are the electrons to the center and the larger effect the centered impurity acts on the electrons. Since the electrons with lower angular momentum states are closer to the donor impurity, the donor and the electrons are under stronger attraction. The state $L=1$ is, therefore, energetically more stable and becomes the lowest state at higher ω_o. In the following, with the combined effect of both impurity and confining strength of $\omega_o=0.06\omega_{LO}$ holding three electrons together in quantum dot, the low-lying states are presented as functions of magnetic fields.

Without the impurity effect, the low-lying states of the three-electron quantum dot for both unpolarized ($S_z=1/2$) and polarized ($S_z=3/2$) states as functions of magnetic fields

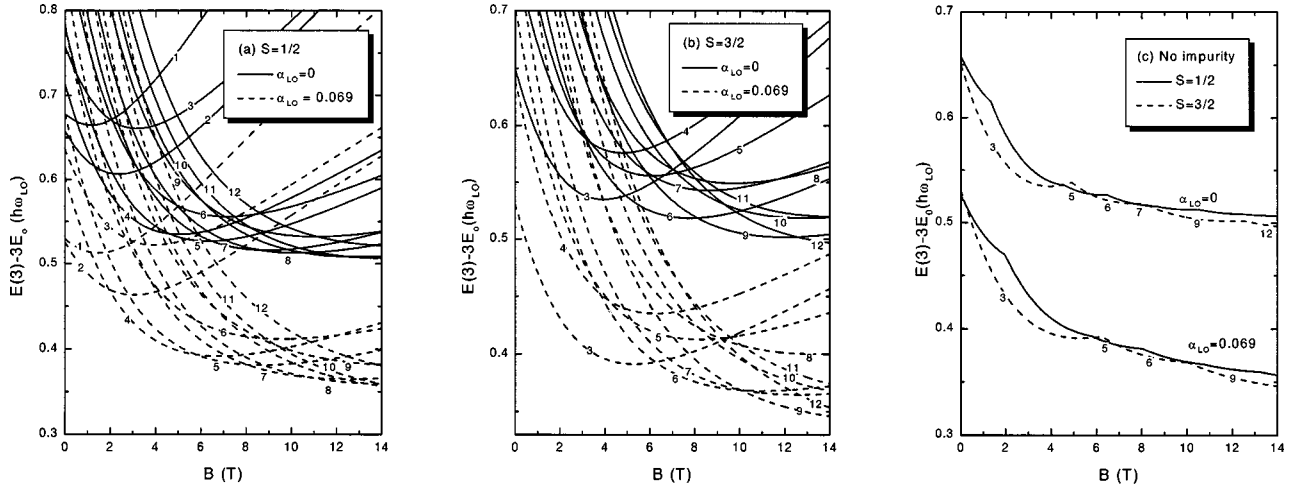

FIG. 2. Lowest-energy states of total orbital angular momentum L with and without polaron effect as functions of magnetic field, with different spin configurations, (a) the total spin $S=1/2$ and (b) the total spin $S=3/2$, and (c) both spin configuration presented together for comparision. Note that $E_o=\hbar\omega_B$.

are plotted in Fig. 2. The low-lying states without and with polaron effects are denoted by solid and dashed lines, respectively. Regardless of the polaron effect, the evolution of the ground state, in general, occurs only at the nonmagic values of total angular momenta $L\neq 3k$, where k is an integer, for unpolarized state and at the magic values $L=3k$ for polarized state. Figure 2(c) presents both spin configurations together for comparison. Overall, when only the Coulomb effect is considered, as the magnetic field is increased from zero, the ground-state transitions occurs in the sequence of $L=3\rightarrow 5\rightarrow 6\rightarrow 7\rightarrow 9\rightarrow 12\cdots$.

When combined with polaron effect, Eq. (A4) contains the square of the sum of three phase factors of the electrons in quantum dot. After expanding, the resulting nine negative phonon induced terms are generally divided into two different types, three self-square and six cross terms. Each of the self-square terms is in fact the self-energy of the corresponding polaron. They are all negative values, which indicate that all three electrons are self-trapped or bound states due to polarization by their Coulomb interactions with the surrounding ions of the lattice. The other six cross terms are also negative which denote that the electrons are attractive via the surrounding strain field, just like Cooper pairs in superconductors. It is worth mentioning here that, if the Frohlich or phonon induced interaction is applied to systems with particles of opposite charges, such as excitons, the cross terms will be positive and the particles will be repulsive. For the present case of a three-electron quantum dot, by comparing the solid and dashed curves in Figs. 2(a) and 2(b), the ground-state energy decreases rapidly as the magnetic-field increases due to the polaron effect. This effect on decreasing the energy of the system are similar for both accepter and donor doped impurities, as depicted by the results shown in Figs. 3 and 4, respectively. As clearly seen in Fig. 2(c), the combined effect of both Coulomb and polaron for every ground-state L/S transition is qualitatively the same as those when only the Coulomb effect is considered, except that the transition shifts slightly towards higher magnetic fields. As the magnetic-field increases, the dot size a decreases and the electrons will in turn jump to the orbits with higher angular

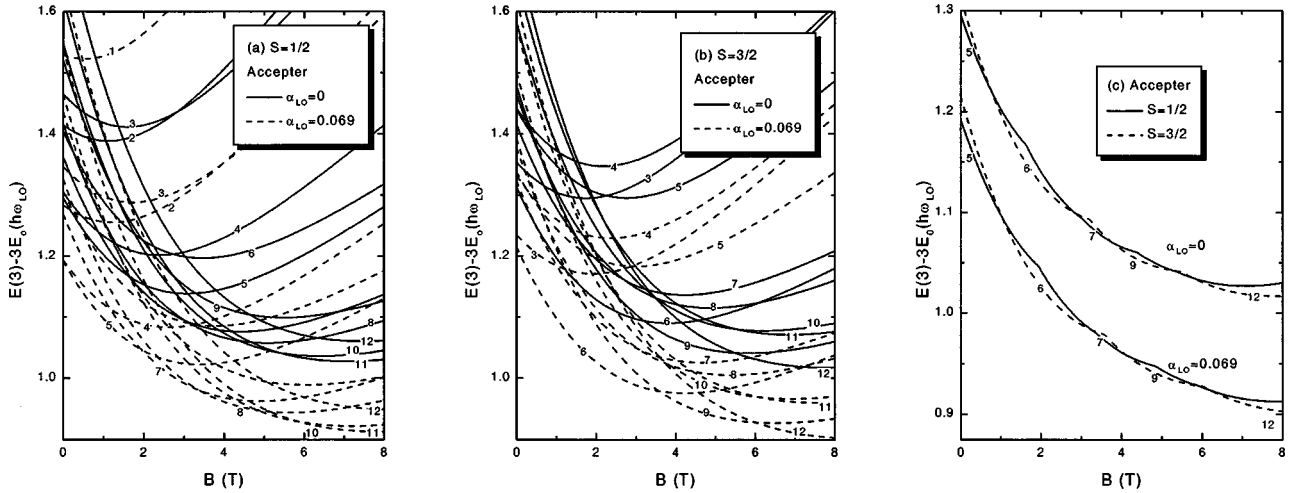

FIG. 3. Same as those in Fig. 2, but with an accepter impurity.

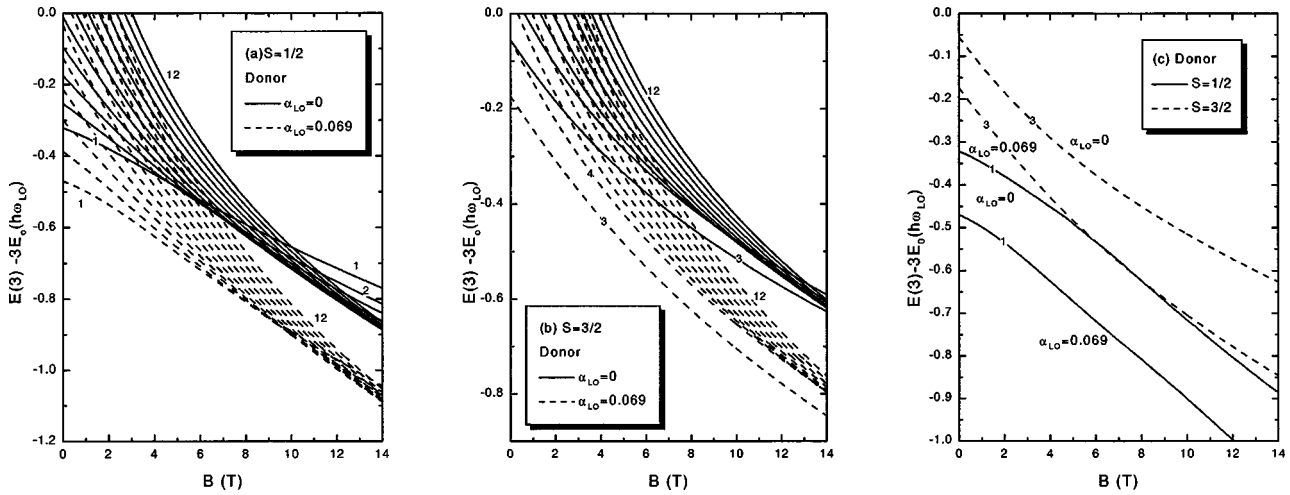

FIG. 4. Same as those in Fig. 2, but with a donor impurity.

momenta to avoid the increase in repulsive energy. When polaron effect is enforced, the electrons are self-trapped and become less repulsive. They jump to higher angular momenta only at higher magnetic fields. Furthermore, the unpolarized state $L=7$ is no longer ground state [see Fig. 2(c)].

In Fig. 3, for an on-center accepter doped quantum dot, similar to that without impurity, the ground-state transits from one angular momentum to another as the magnetic-field increases. Since the accepter provides additional repulsion to the electrons, they have to jump to higher states even in lower magnetic fields in order to minimize the energy of the whole system. The electrons at lower angular momenta are closer to the center of the quantum dot and are energetically unstable because of the electron-accepter repulsion. They all repel from the center of the quantum dot, as we see in Fig. 3(a)for unpolarized states, $l=2,4$ and in Fig. 3(b)for polarized states $l=3$. They are no longer ground states. Furthermore, the ground-state energies of the three-electron quantum dot for both unpolarized and polarized states shift $0.6\hbar\omega_{LO}$ higher comparing with those without impurity.

In Fig. 4, the low spectra for on-center donor doped quantum dot are presented. It is clearly observed that there are, in general, no crossovers of ground-state orbital angular momenta, except unclear level crossing of angular momentum $L=1$ for unpolarized state with higher ones at higher magnetic fields, $B>6T$. Since there are two interaction energies, electron-electron repulsion and electron-donor attraction, competing with each other, and the electron-donor attraction is much stronger, the electrons are much closer to the center of the dot and become localized in the vicinity of the center. Therefore, the electrons do not jump to higher angular momenta or separate from the on-center donor impurity even at very high magnetic fields.

The transition of orbital L or spin S angular momentum in ground-state energy can be appeared as discontinuities in the magnetic properties of the quantum dot since they are clear-cut consequence of resulting interaction of the whole system. The magnetic moment μ_{mag} and the magnetic susceptibilty χ can be defined as $\mu_{mag}=-\partial E_{\mathrm{ground}}(3)/\partial B$ and $\chi=\partial\mu_{mag}/\partial B$, respectively. In Fig. 5, the magnetic moments

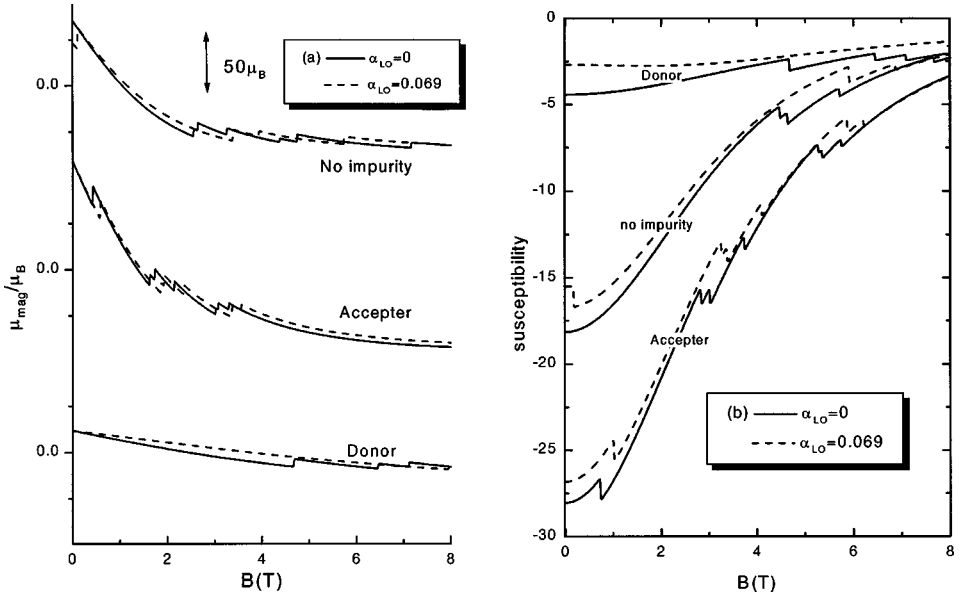

FIG. 5. (a) Magnetic moment μ_{mag} in units of Bohr magneton μ_B and (b) magnetic susceptibility of a three-electron quantum dot.

and the magnetic susceptibility of the three-electron quantum dot with (dashed lines) and without (solid lines) polaron effect are shown. With polaron effect, the spikes of both magnetic moment and the susceptibility shift to higher magnetic fields.

In summary, a numerical method for the calculation of the eigenenergies of N electrons bound to quantum dots with Coulomb impurity is developed. As an example, using numerical diagonalization, we calculate the energy spectra of a three-electron polar quantum dot with and without Coulomb impurity. Three remarks on the numerical results are worth noting.

(1) There is a clear-cut transition at $\omega_o \approx 0.035\omega_{LO}$ without polaron effect and $0.025\omega_{LO}$ with polaron effect for the donor doped quantum dot as the confining strength increases.

(2) In the presence of electron-phonon interaction, the S/L ground-state transitions shift towards higher magnetic fields with respect to the case that only the Coulomb effect is considered.

(3) For both magnetic moment and magnetic susceptibilty, there are ground-state L/S transitions or spikes for quantum dots with or without centred acceptor. However, the ground-state transitions are generally suppressed for those with donor impurity.

The phonon induced Hamiltonian derived in the present study can be applied to systems with more than three electrons and even those with particles of opposite charges, such as excitons. The present formalism provides reference to calculate the energy spectra of the few body systems involving excitons or charged excitons, which is of growing interest.[48] The present results also motivate further study on the effect of the position of the impurity, inside and outside of a few-electron system.

ACKNOWLEDGMENTS

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APPENDIX

If the whole set of quantum numbers is denoted by the subscript $[k]$ in brevity, in the case of three electrons, the matrix elements $\langle \psi_{[k]}|\hat{H}|\psi_{[k']}\rangle$ are given by

$$\langle \psi_{[k]}|\hat{H}_0|\psi_{[k']}\rangle = \Big\{ [2(n_1+n_2+n_3) + |l_1|+|l_2|+|l_3|+3]\hbar\omega_B + \frac{1}{2}\hbar\omega_c(l_1+l_2+l_3) + g^*\mu_B B S_z \Big\} \delta_{[k],[k']}, \tag{A1}$$

$$\langle \psi_{[k]}|\hat{H}_{\rm co}|\psi_{[k']}\rangle = \frac{e^2}{\epsilon_\infty}\sum_{i<j}\langle \psi_{[k]}|\frac{1}{|\vec{\rho}_i-\vec{\rho}_j|}|\psi_{[k']}\rangle, \tag{A2}$$

$$\langle \psi_{[k]}|\hat{H}_{\rm imp}|\psi_{[k']}\rangle = \frac{e^2}{\epsilon_\infty}\sum_{i=1}^{3}\langle \psi_{[k]}|\frac{1}{\rho_i}|\psi_{[k']}\rangle, \tag{A3}$$

$$\langle \psi_{[k]}|\hat{H}_{\rm ind}|\psi_{[k']}\rangle = -\frac{\alpha_{LO}\hbar\omega_{LO}}{u_{LO}}\int_0^\infty dq_\parallel \langle \psi_{[k]}|\sum_{i=1}^{3}\zeta_i e^{i\vec{q}\cdot\vec{\rho}_i}|\psi_{[k']}\rangle|^2, \tag{A4}$$

where u_{LO} is the polaron size and $\zeta_i = 1$ for all three electrons. In the numerical work, for a particular total orbital momentum L and particular z-component spin of the total electrons' spins of the whole system, the whole sets of bases with the total energies $2(n_1+n_2+n_3)+|l_1|+|l_2|+|l_3|$ including all combinations of the z component of electron spins are first arranged in an increasing order, before performing the numerical diagonalization for $S_z = 1/2$ and 3/2, respectively. The accuracy of the numerical solution depends on the size of the model space. No more than 42 bases are involved in the present diagonalization. Note that the z component S_z can take the values of $S, S-1, \ldots, -S$ for given S. In other words, for the final two sets of eignenergies, $S_z = 1/2$ and 3/2, those low-lying states of $S_z = 1/2$ belong to the spin state $S = 3/2$, whereas the remaining low-lying states of $S = 1/2$ belong to $S = 1/2$.

*FAX: 852-2788-8423; Email address: mejyau@cityu.edu.hk
†Email address: mesimon@cityu.edu.hk

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