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Spin polarization of electrons in GaAs quantum dots: role of electron correlations

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Abstract. The ground state behaviour of GaAs quantum dots with $N = 2-6$ electrons in a magnetic field of $B = 0-5$ T is investigated in the Hartree–Fock (HF) approximation, and then the correlation effects are taken into account via a local-density-functional approach. It is found that in the HF approximation the dot electrons have already been in spin-polarized states at small B , even at $B = 0$ T for most cases. However when the electron correlations are included, all the spin-polarized states at small B are suppressed and the ground states of all cases around $B = 0$ are normal states with $S = 0$ or $\frac{1}{2}$ for even or odd number of electrons. As B is increased to 1.83–2.64 T the ground states will directly transit from normal states to their corresponding fully spin-polarized states.

As is known, due to the low effective density and the restriction of electron motion to only two dimensions, the electrons in quantum dots are usually highly correlated. To fully understand the experimental results and to correctly predict the unknown properties of quantum dots, the correlation effects must be considered carefully. The existing theoretical approaches to solve the electronic structures of quantum dots mainly include the exact numerical diagonalization [1, 2] and the Hartree–Fock (HF) approximation [3–5]. Though the exact numerical diagonalization treatment can completely include the correlation effects, it is limited by the convergence problems to treating just a few electrons. This is because electron correlations in quantum dots are relatively much stronger than in atoms and molecules, to a certain accuracy, the size of the basis set used to numerically diagonalize the Hamiltonian increases rapidly with the electron number; this makes the numerical calculation exceedingly difficult for more than six electrons [6]. The HF method, however, is potentially applicable to more electrons but, because of neglect of the electron correlation, its accuracy is doubtful. The theoretical study of Pfannkuche *et al* [7] on a two-electron quantum dot has shown that the HF approximation is significantly in error and can even give qualitatively incorrect results about the spin singlet–triplet transition of the two electrons [8, 9], which has been observed experimentally [10]. For the case of three spin polarized electrons in a strong magnetic field, the comparison of the HF and the exact numerical diagonalization results shows that the correlation effects on the ground state are very small but those on the low-lying energy level structure are apparent [11, 12].

In this paper, we shall systematically study the important role of electron correlations in the process of spin polarization of quantum dot electrons in a magnetic field. To do so, the calculations are performed first in the HF approximation, and then the correlation effects are included via a local-density-functional approach. We should mention that the local-density-functional approach is an approximate way to include the electron correlations and

has its own limitations [13], but it does not suffer from computational difficulty as in the exact numerical diagonalization treatment and can correctly give the differences between the correlation energies of different spin polarized states, which are very important for the present study. This work can be considered as an extension of the study of Pfannkuche *et al* [7], but in an alternative approach.

Since it is believed that the dots currently studied experimentally confine the electrons by a potential that is quadratic to a good approximation when the electron number N is small [1], so the present calculations, as most previous theoretical studies, have been done for the case of ideally two-dimensional electrons, in a circular dot in the x - y plane, confined by a parabolic potential with a magnetic field B along the z axis. The total Hamiltonian can be written as

$$\hat{H} = \sum_{i=1}^N \hat{h}(i) + \frac{e^2}{4\pi\epsilon\epsilon_0} \sum_{i<j} \frac{1}{r_{ij}} \quad (1)$$

where

$$\hat{h}(i) = \frac{1}{2m^*} (\mathbf{p}_i + e\mathbf{A}_i)^2 + \frac{1}{2} m^* \omega_0^2 r_i^2 + \frac{g^* \mu_B}{\hbar} B \hat{s}_{i,z} \quad (2)$$

is the single-electron Hamiltonian. Here m^* is the electron effective mass, g^* is the effective g factor, μ_B is the Bohr magneton, $\hat{s}_{i,z}$ is the z component of the spin operator of electron i , and $\mathbf{A}_i = (B/2)(-y_i, x_i, 0)$ the vector potential in the symmetric gauge.

In the HF approximation, the many-electron problem is reduced to a one-electron problem. An one-electron wavefunction or a spin orbital is constructed as a simple product of a spatial orbital and a spin function, and a Slater determinant can be constructed by using N spin orbitals.

Since the eigenstates of the system are the eigenstates of all three operators \hat{L}_z , \hat{S}^2 and \hat{S}_z , which are the total angular momentum operator, the square operator of total spin and its z component respectively, they can be classified by quantum numbers M ($= \sum_{i=1}^N m_i$), S and M_S ($= \sum_{i=1}^N m_{s_i}$). In general, if an N -electron determinant has n_α columns with α spin and n_β columns with β spin ($n_\alpha \geq n_\beta$), the determinant will not be an eigenfunction of \hat{S}^2 unless the number of doubly occupied spatial orbitals is equal to n_β . Fortunately, the energy states we are interested in here are just the lowest ones for given values of S , which, in the presence of a magnetic field, correspond to $M_S = S$. This requires that among the N spin orbitals (n_i, m_i, m_{s_i}) used to construct the Slater determinant all the singly occupied spatial orbitals should be occupied by electrons with α spin, so that the determinant must be the eigenfunction of \hat{S}^2 . Therefore, the following calculations can be carried out in the framework of one determinant HF method.

As in [11] and [12], we also expand the spatial parts of the one-electron wavefunctions in a set of Gaussian functions, that is

$$\psi_{nm}(\mathbf{r}) = \sum_p C_{nmp} \chi_{mp}(\mathbf{r}) \quad (3)$$

where

$$\chi_{mp}(\mathbf{r}) = \frac{2^{(|m|+2)/2} \alpha_p^{(|m|+1)/2}}{(|m|!)^{1/2}} r^{|m|} e^{-\alpha_p r^2} \frac{1}{\sqrt{2\pi}} e^{-im\varphi}. \quad (4)$$

By choosing some finite basis set size to minimize the Hamiltonian integral and varying the expansion coefficients C_{nmp} for a fixed set of exponents α_p , we obtain the HF secular equations [14]

$$\mathbf{FC} = \epsilon \mathbf{SC} \quad (5)$$

where the overlap matrix \mathbf{S} and the Fock matrix \mathbf{F} are defined by

$$S_{mpq} = \langle \chi_{mp} | \chi_{mq} \rangle \quad (6)$$

$$F_{mm_s pq} = h_{mpq} + G_{mm_s pq} \quad (7)$$

$$h_{mpq} = \langle \chi_{mp} | \hat{h} | \chi_{mq} \rangle \quad (8)$$

$$G_{mm_s pq} = \sum_{n', m', m'_s}^{occ} \sum_{r, s} C_{n'm'r}^* C_{n'm'_s} [(mpmq | m'rm'_s) - \delta_{m_s, m'_s} (mpm'_s | m'rmq)] \quad (9)$$

with

$$(mpmq | m'rm'_s) = \frac{e^2}{4\pi\epsilon\epsilon_0} \langle \chi_{mp}(1) \chi_{m'r}(2) \left| \frac{1}{r_{12}} \right| \chi_{mq}(1) \chi_{m'_s}(2) \rangle. \quad (10)$$

Solving equation (5) iteratively, we can obtain the spin-orbital energies ϵ_{nmm_s} , which are related to the total energy E^{HF} by

$$E^{HF} = \frac{1}{2} \left[\sum_{n, m, m_s}^{occ} \epsilon_{nmm_s} + \sum_{n, m, m_s}^{occ} \sum_{p, q} C_{nmp}^* C_{nmq} h_{mpq} \right]. \quad (11)$$

The summation \sum_{n, m, m_s}^{occ} in equations (9) and (11) is over all occupied spin orbitals. For given values of M and S , the lowest state is labelled as $|OMS\rangle$, which is in the HF approximation a Slater determinant constructed from a set of occupied spin orbitals (n_i, m_i, m_{s_i}) satisfying the conditions $M = \sum_{i=1}^N m_i$ and $S = M_s = \sum_{i=1}^N m_{s_i}$, and giving the lowest total energy.

The correlation corrections to the HF energies of the lowest states with given values of M and M_s are taken into account via the local density approximation (LDA) in which the functional form of the correlation energy $E_c(r_s, \xi)$ for a two-dimensional electron gas given by Tanatar and Ceperley [15] is used. The dimensionless density parameter is defined as $r_s = a/a_0^*$, where a_0^* is the effective Bohr radius and $a = 1/\sqrt{\pi\rho}$ is the radius of the circle that encloses one electron on average and ρ is the number density. The parameter ξ is the degree of polarization. Since the parameters in the expression of $E_c(r_s, \xi)$ are known only for the cases of $\xi = 0$ and 1, the correlation energy for the case of $0 < \xi < 1$ is estimated by a linear interpolation, that is, $E_c(r_s, \xi) = (1 - \xi)E_c(r_s, 0) + \xi E_c(r_s, 1)$. The number density ρ is determined using HF wavefunctions. Though this is not a self-consistent way to calculate the correlation energies, the results should be reasonable because the HF wavefunctions are very close to the exact ones.

The lowest energy levels for given values of M and S of quantum dots with $N = 2-6$ electrons varying with the magnetic field strength B are plotted in figure 1 to figure 5. They have been calculated using parameters appropriate to GaAs ($m^* = 0.067 m_0$, $\epsilon = 12.4$ and $g^* = -0.44$). The confining energy $\hbar\omega_0$ is set to be 3.37 meV, which corresponds to a dot with characteristic length $l_0 = (\hbar/m^*\omega_0)^{1/2} = 18.4$ nm, so that the present result for two electrons can be compared with that in [6]. Since we are interested only in the process of spin polarization, so the magnetic field is limited below 5 T, above which the electrons are always completely spin polarized. To show the results clearly, the curves of HF and HF plus correlations are plotted separately for $N = 4-6$. From these figures, we can find that in the HF approximation the spin-polarized states are favourable in energy. Even at $B = 0$, the HF ground states are spin-polarized states except for a two-electron dot, in which a magnetic field larger than 0.15 T, as shown in figure 1, is needed to keep the two electrons spin polarized. For $N = 4$ (figure 3(a)), when B is smaller than 0.10 T, the HF ground state is $|001\rangle$, a partially spin-polarized state with $S = 1$, and as B is increased, a spin-multiplet transition from $|001\rangle$ to $|022\rangle$ will occur, the four electrons thus being fully spin polarized. The situation for $N = 5$ (figure 4(a)) is a little more complicated: a partially spin-polarized HF ground state $|023/2\rangle$

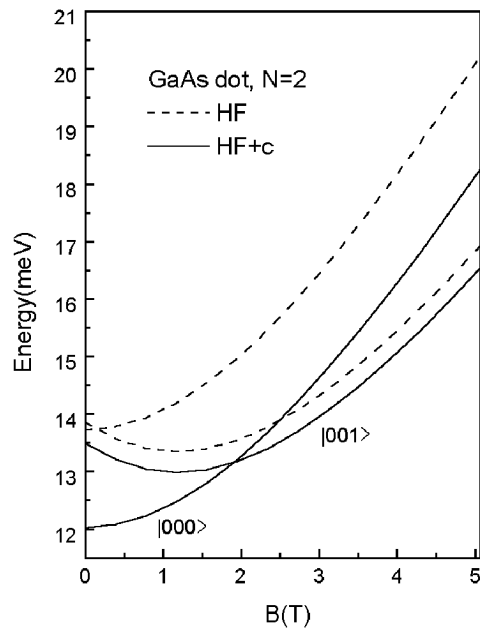


Figure 1. The lowest energy levels for given values of M and S of a two-electron GaAs quantum dot as a function of the magnetic field.

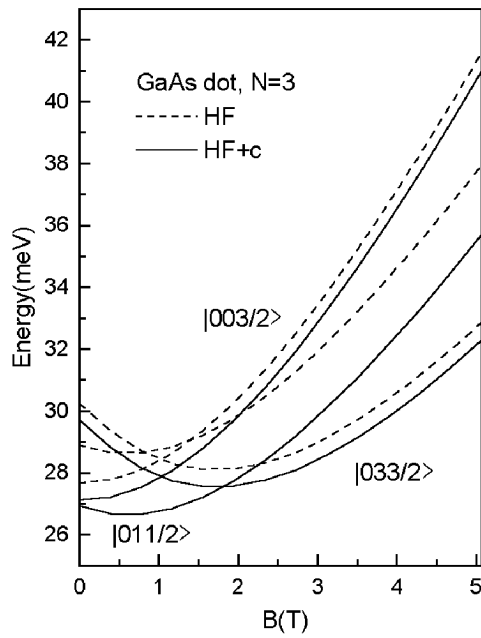


Figure 2. The lowest energy levels for given values of M and S of a three-electron GaAs quantum dot as a function of the magnetic field.

appears in the region between $B = 0.27$ and 0.75 T. This means that the ground state will experience spin-multiplet transitions from $|005/2\rangle$ to $|023/2\rangle$ and then from $|023/2\rangle$ to

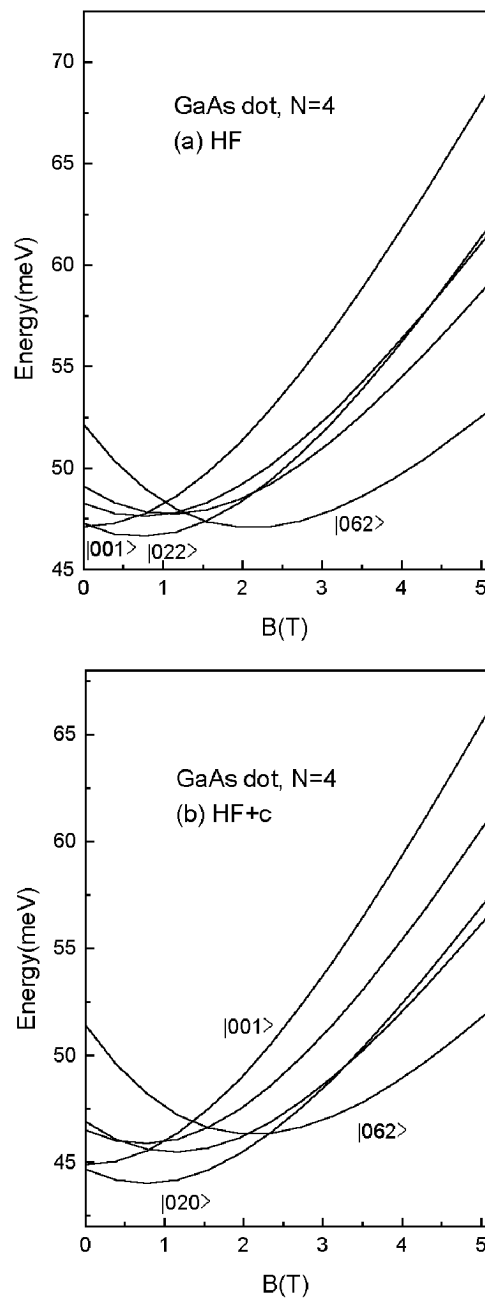


Figure 3. The lowest energy levels for given values of M and S of a four-electron GaAs quantum dot as a function of the magnetic field. (a) HF approximation, the plotted levels at $B = 0$ T from bottom to top: $|0MS\rangle = |001\rangle, |022\rangle, |020\rangle, |031\rangle, |062\rangle$; (b) HF approximation plus electron correlations, the plotted levels at $B = 0$ T from bottom to top: $|0MS\rangle = |020\rangle, |001\rangle, |022\rangle, |031\rangle, |062\rangle$.

$|055/2\rangle$. The appearance of this phenomenon may attribute to the competition between the exchange energy and the additional energy in the magnetic field. For $N = 3$ (figure 2) and 6

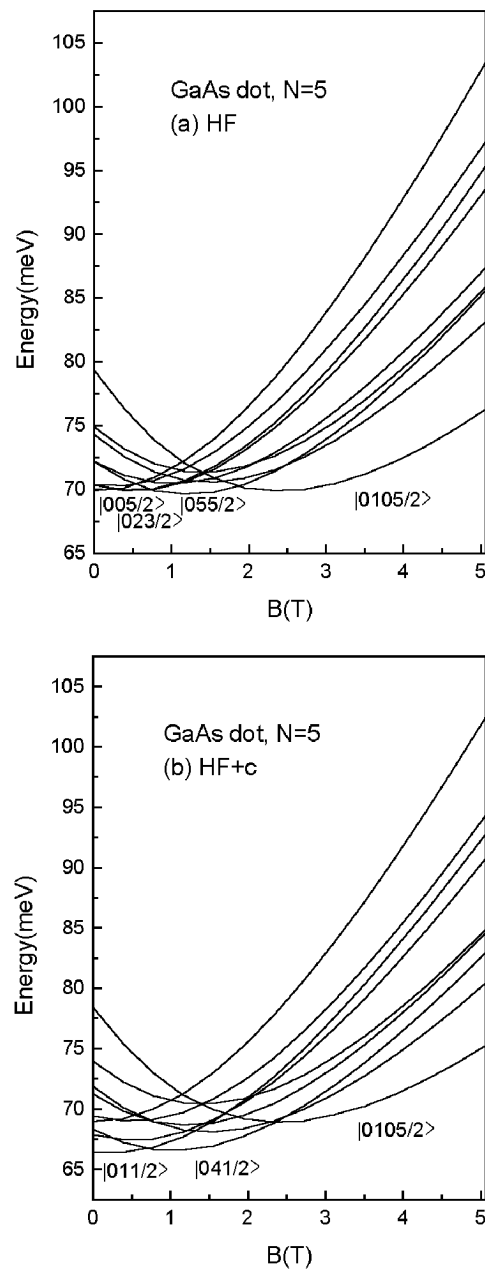


Figure 4. The lowest energy levels for given values of M and S of a five-electron GaAs quantum dot as a function of the magnetic field. (a) HF approximation, the plotted levels at $B = 0$ T from bottom to top: $|0MS\rangle = |005/2\rangle, |023/2\rangle, |025/2\rangle, |011/2\rangle, |041/2\rangle, |055/2\rangle, |063/2\rangle, |065/2\rangle, |0105/2\rangle$; (b) HF approximation plus electron correlations, the plotted levels at $B = 0$ T from bottom to top: $|0MS\rangle = |011/2\rangle, |023/2\rangle, |041/2\rangle, |005/2\rangle, |025/2\rangle, |055/2\rangle, |063/2\rangle, |065/2\rangle, |0105/2\rangle$.

(figure 5(a)), the HF ground states are always fully spin-polarized states at any value of B , and there only exist ground state angular momentum transitions as B is increased.

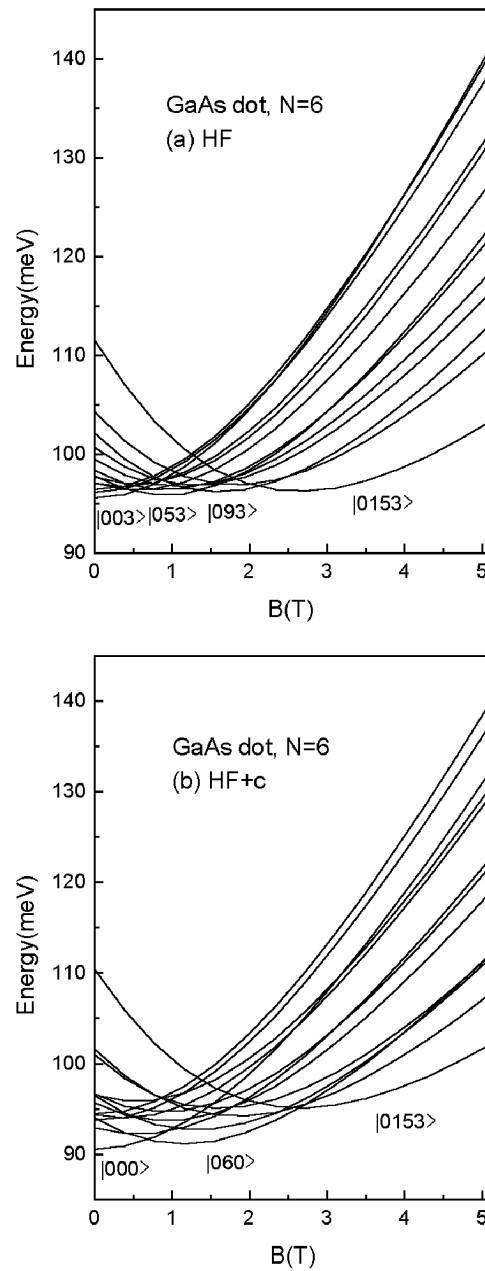


Figure 5. The lowest energy levels for given values of M and S of a six-electron GaAs quantum dot as a function of the magnetic field. (a) HF approximation, the plotted levels at $B = 0$ T from bottom to top: $|0MS\rangle = |003\rangle, |000\rangle, |002\rangle, |022\rangle, |031\rangle, |033\rangle, |053\rangle, |052\rangle, |060\rangle, |071\rangle, |093\rangle, |0102\rangle, |0153\rangle$; (b) HF approximation plus electron correlations, the plotted levels at $B = 0$ T from bottom to top: $|0MS\rangle = |000\rangle, |031\rangle, |002\rangle, |060\rangle, |022\rangle, |003\rangle, |052\rangle, |033\rangle, |071\rangle, |053\rangle, |093\rangle, |0102\rangle, |0153\rangle$.

As illustrated in figure 1 to figure 5, when the correlation energies are included the situation changes a great deal. Instead of the spin-polarized states, the normal states with $S = 0$ (for

even N) or $S = 1/2$ (for odd N) become favourable when B is small. As B is increased to certain values, 1.90, 1.83, 2.30, 2.37 and 2.64 T for $N = 2, 3, 4, 5$ and 6 respectively, the ground states will directly transit from normal spin states to fully spin-polarized states, no partly spin-polarized states being observed. The reason for the lower transition value of B for $N = 3$ is not very clear. We can also see that, before the transitions to fully spin-polarized states occur, the ground state angular momenta do not change for $N \leq 4$, but for $N = 5$ and 6 the ground state angular momenta will transit from $M = 1$ to 4 and from $M = 0$ to 6, respectively, both at about $B = 0.70$ T.

The main reason to lead the spin-polarized states to be favourable in the HF approximation, especially at $B = 0$, is that for the states with larger S (thus with larger M_s) the exchange energies are larger in magnitude. But in contrast, for the states with larger S (thus with larger ξ) the correlation energies are smaller in magnitude. The above results show that in the competition between the exchange and correlation interactions at small B , the latter gets the upper hand, so the spin polarizations are suppressed until B becomes large enough. Indeed, since the work of Pfannkuche *et al* [7], the important role of electron correlations in determining the spin-multiplet states of quantum dot electrons had been clearly realized. The present and Pfannkuche *et al* results for $N = 2$ agree well with each other, the present value of B , 1.90 T, at which the spin singlet–triplet transition occur being very close to that of Pfannkuche *et al*, 1.95 T (estimated from the figure published). By comparing with the result of Pfannkuche *et al* we also find that the present correlation energies in the LDA are a little overestimated, for example, in the state $|000\rangle$ at $B = 1$ T the correlation energies of two GaAs quantum dot electrons in the present and Pfannkuche *et al* calculations are -1.72 and -1.43 meV, respectively. But fortunately, this situation is also true for almost all the spin-multiplet states, therefore, the errors of present approximation do not affect significantly the ground state behaviour of the dot electrons, and as the number of electrons is increased, the LDA will work better. So, we have reason to believe that the present results are reliable.

To summarize, the important role of electron correlations in determining the spin-multiplet states of GaAs quantum dots with up to six electrons is investigated by comparing the ground state behaviours of the dot electrons before and after the correlation energies are included via the LDA. The results show that, because of electron correlation interactions, the phenomenon of spin polarization of dot electrons appearing in the HF approximation at a small B disappears, and a magnetic field of $B = 1.83$ – 2.64 T is needed to keep two to six electrons in GaAs quantum dots spin polarized. This indicates again that HF approximation is qualitatively incorrect in predicting the spin-multiplet structures of quantum dot electrons at small B and the electron correlation effects cannot be neglected in treating the problems concerning the spin-multiplet transitions of dot electrons. Though the present study is only done for up to six electrons, the study for more than six electrons is just a dull routine work in the same way.

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