Spontaneous Magnetization and Electron Momentum Density in 3D Quantum Dots

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We discuss an exactly solvable model Hamiltonian for describing the interacting electron gas in a quantum dot. Results for a spherical square well confining potential are presented. The ground state is clearly found to exhibit spontaneous magnetization, indicating that this effect would be amenable to direct experimental observation via electron momentum density spectroscopies such as angular correlation of positron annihilation radiation (ACAR) and Compton scattering. Our results also bear more broadly on the applications of quantum dots in that they point to the possibility of obtaining a system in a ferromagnetic ground state without requiring low temperatures and/or external magnetic fields.

We consider the Hamiltonian

\[ \hat{H} = \sum_{\nu \sigma} \epsilon_{\nu} a_{\nu \sigma}^\dagger a_{\nu \sigma} + \frac{1}{2} U \sum_{\nu' \nu \sigma' \nu' \sigma} a_{\nu \sigma}^\dagger a_{\nu' \sigma'}^\dagger a_{\nu' \sigma'} a_{\nu \sigma}. \]  

The first term (\( \hat{H}_0 \)) describes the noninteracting system. It corresponds to the spherical square well: \( V(r) = -V_0 \), for \( r \leq R \), and \( V(r) = 0 \) otherwise, and the associated one-particle eigenfunctions \( \phi_\nu \), with eigenvalues \( \epsilon_\nu \). \( \nu \equiv nlm \) is a composite index and \( \sigma \) labels spin. The second term (\( \hat{H}_1 \)) describes the interaction and allows Coulomb repulsion via the parameter \( U \) only between electrons of opposite spin—electrons of like spin being kept apart by the Pauli exclusion principle. In this connection, we have carried out extensive numerical estimates, and find that direct Coulomb matrix elements are larger, on the average, by an order of magnitude or more, than exchange or other terms for dot radii up to 30 Å and electron densities ranging from \( r_s = 2 \) to 5 (Bohr radii) [13]. Spin-orbit effects here are also expected to be small, since the confining potential is relatively weak due to the screening of the nuclei by the core electrons. For these reasons, form (1) should provide a reasonable model for the electronic structure of 3D quantum dots.

The standard one-particle Green function for the many electron system can be expanded as

\[ G_{\sigma \sigma'}(r r', \tau) = \sum_{\nu \nu'} \varphi_\nu^\dagger(r) \varphi_{\nu'}(r') G_{\sigma \sigma'}(\nu \nu', \tau), \]

with

\[ G_{\sigma \sigma'}(\nu \nu', \tau) = -\langle T_\tau a_{\nu \sigma}(\tau) a_{\nu' \sigma'}^\dagger(0) \rangle. \]

It is readily shown that \( \hbar \partial_{\nu} / \partial \tau = -\xi_\nu a_{\nu \sigma} - U N_{- \sigma} a_{\nu \sigma} \), where \( \xi_\nu = \epsilon_\nu - \mu \) (\( \mu \) is the chemical potential), and \( \hat{N}_{- \sigma} = \sum_{\nu} a_{\nu \sigma}^\dagger a_{\nu \sigma} \). Then, the equation of motion for the Green function \( G_{\sigma \sigma'}(\nu \nu', \tau) \) is

\[ -\hbar \frac{\partial}{\partial \tau} G_{\sigma \sigma'}(\nu \nu', \tau) = \hbar \delta_{\sigma \sigma'} \delta_{\nu \nu'} \delta(\tau) + \xi_\nu G_{\sigma \sigma'}(\nu \nu', \tau) \]

\[ - U \langle T_\tau \hat{N}_{- \sigma}(\tau) a_{\nu \sigma}(\tau) a_{\nu' \sigma'}^\dagger(0) \rangle. \]
Since our Hamiltonian possesses the property that $[H, \hat{N} - \mu N] = 0$, Wick’s theorem allows us to factorize the preceding ground state average, i.e.,

$\langle \hat{T}_s \hat{N}_{-\sigma}(\tau) a_{v\sigma}(\tau) a^\dagger_{v\sigma}(0) \rangle = -N_{-\sigma} \mathcal{G}_{\sigma\sigma'}(\nu \nu', \tau)$, where $N_{-\sigma} = \sum_{v'} f_{v' - \sigma}$, with $f_{v' - \sigma}$ denoting the Fermi occupation function. A Fourier transform in $\tau$ immediately yields the exact Green function

$$G_{\sigma\sigma'}(\nu \nu', i\omega_n) = \delta_{\sigma\sigma'} \delta_{\nu\nu'} \frac{1}{i\omega_n - \xi_{\nu}/\hbar - U N_{-\sigma}/\hbar}. \quad (3)$$

The self-energy thus is $\Sigma_\sigma = U N_{-\sigma}/\hbar$. The interacting energy levels, $\epsilon_{v\sigma}$, are given by the solutions of $\omega - \xi_{\nu}/\hbar - \Sigma_\sigma = 0$ (frequencies measured with respect to $\mu/\hbar$ minimizing the ground state energy, $E = \sum_{v\sigma} (\epsilon_{v\sigma}^0 + \frac{1}{2} \Sigma_\sigma^2) f_{v\sigma}$. Given the number of particles $N$, this defines a set of nonlinear equations for the populations, $N_{\uparrow}$ and $N_{\downarrow}$, of up and down spin states, respectively. The resulting splitting in energy for states of opposite spin, $\Delta = U(N_{\uparrow} - N_{\downarrow})$, is uniform, i.e., it does not depend on the quantum numbers $\nu$. Note that temperature enters our formulae only through the Fermi function, $f_{v\sigma}$; all results in the remainder of this article refer to the zero temperature limit.

We comment briefly on the nature of the four essential parameters which describe our model: $r_s$, the electron density; $R$, the dot radius or, equivalently, the number $N$ of electrons [$N = (R/r_s)^3$]; $V_0$, the well-depth; and, $U$, the Coulomb energy. The values of $r_s$ and $R$ are related to the type and size of the systems considered. In this study we use $r_s = 5$, a relatively low density, enabling us to consider a wide range of dot radii. $V_0$ may then be obtained reasonably in terms of the work function of the system studied and the Fermi energy of the free electron gas with the same $r_s$. For $r_s = 5$, following Ref. [15], we take $V_0 = 8.62$ eV. The choice of the remaining parameter $U$ is somewhat tricky, mainly because correlations in quantum dots are not well understood. A handle on $U$ can be obtained by considering the average value $U_{TF}$ of the Thomas-Fermi (TF) screened Coulomb interaction, $\int d\mathbf{r} d\mathbf{r}' |\phi_\nu(\mathbf{r})|^2 v_{TF}(|\mathbf{r} - \mathbf{r}'|)|\phi_\nu(\mathbf{r}')|^2$, averaged over all occupied states in the noninteracting system, with $v_{TF}(r) = e^2 \exp(-r/l_{TF})/r$, where $l_{TF} = \sqrt{\epsilon_{\nu}/1.56}$ is the TF screening length. However, actual screening in real materials is likely to be weaker. For example, in noble metals, Ref. [16] reports an effective screening length $l_{TF}/0.73$. Using this value, we find an enhanced Coulomb interaction given roughly by $1.75 U_{TF}$, which is the value we have used throughout this work.

Fig. 1 illustrates the nature of the energy levels $\epsilon_{v\sigma}$ for a 12.11 Å radius dot, for which $N = 96$, and $U = 27.13$ meV ($U_{TF} = 15.50$ meV). The self-consistent splitting between up and down spin electrons is found to be, $\Delta = 1.03$ eV. The highest occupied molecular orbital (HOMO) energy level is, $E_{\text{HOMO}} = -5.86$ eV, which may be thought of as the dot “Fermi energy”. We have $N_{\uparrow} = 67$ and $N_{\downarrow} = 29$, so that the ground state spin polarization per electron is $\zeta = (N_{\uparrow} - N_{\downarrow})/N = 0.4$.

We discuss now $\zeta$ as a function of dot radius with reference to Fig. 2. In the weakly interacting limit ($U \to 0$), $\Delta \to 0$, still the degeneracy between up and down spin in the nl shells is lifted, and as Fig. 2(a) clearly shows, shells fill in accord with Hund’s first rule. Indeed, as the number of particles increases with dot radius, the spin polarization reaches a peak each time a shell is half filled with up-spin electrons, and falls to zero when the shell is completed with down-spin electrons. The sequence of these so-called “magic numbers”, i.e., $N$ values for which all occupied shells are completely filled, will of course differ here from that in atoms or 2D quantum dots due to the presence of a finite dot size.
to differences in the details of the underlying spectrum. The magic numbers up to 106 are indicated on the upper axis in Fig. 2(a). Turning next to the strongly interacting case, Fig. 2(b) shows large deviations in $\zeta$ from a simple Hund’s rule filling [17]. This is because $\Delta$ changes with each added electron in order to minimize the total energy. The first 4 electrons enter the up spin 1$s$ and 1$p$ levels, so that $\zeta = 1$, while the next 4 enter the corresponding down spin levels until $\zeta$ vanishes for $N = 8$. The 9th electron induces a strong change in $\Delta$, so that the 1$s$ and 1$p$ down spin levels are pushed above the 1$d$ up spin level, causing the former to empty in favor of the latter and the system to become completely polarized again. Similar level reorderings are involved in the other “kinks” seen in Fig. 2(b).

In order to identify observable signatures of the remarkable changes in the polarization depicted in Fig. 2, we consider the electron momentum density (EMD), defined by $n(p) = (2\pi)^{-1} \int_{-\infty}^{\infty} d\omega \, f(\omega) A(p, \omega)$, where $f$ is the Fermi function and $A$ is the spectral function $A(p, \omega) = -2 \text{Im} G^R(p, \omega)$, with $G^R(p, \omega) = G(p, i\omega_n \rightarrow \omega + i\delta)$. The inset in Fig. 3 illustrates $n(p)$ and the magnitude of its first derivative, $|n'(p)|$, for the same parameter values as in Fig 1. The region of rapid variation in $n(p)$ can be characterized via the position, $p_F$, of the peak in $|n'(p)|$ and the associated full-width-at-half-maximum (FWHM), $\Delta p$. In the bulk limit in a metallic system, $p_F$ will tend to the Fermi momentum, where $n(p)$ will develop a break and $|n'(p)|$ a $\delta$-function peak, correspondingly. For simplicity, therefore, we may refer to $p_F$ loosely as the dot “Fermi momentum”, even though there are no breaks in the EMD in a finite system [18,19]. Fig. 3 considers $\Delta p$ systematically and shows the presence of a dramatic series of peaks (solid line) spaced more or less regularly as a function of $R$. A comparison with corresponding changes in $\zeta$ (dotted line), makes it evident that peaks in $\Delta p$ are well correlated with those in $\zeta$, some differences notwithstanding. This correlation arises because, at a peak in $\zeta$, the up and down spin EMDs become substantially shifted with respect to each other, reflecting the polarization of the system, so that the EMD presents a much larger value of $\Delta$ overall. In short, peaks in $\Delta p$ clearly are a signature of peaks in $\zeta$.

Motivated by the preceding considerations, we have obtained an approximate expression for $\Delta p$ as

$$\frac{\Delta p}{p_F} = \Delta p_0/p_F + c(\zeta - \zeta_0) R,$$

where $\zeta_0$ is the spin polarization in the weakly interacting case ($U \rightarrow 0$), and $c$ is a constant, which depends on various dot parameters, with a fitted value in the present case of $c = 4.29 \times 10^{-2} \text{eV}^{-1} \text{Å}^{-1}$. The first term in Eq. 4 describes the $U \rightarrow 0$ limit and can be shown to follow the scaling law $0.93 A(r_s/a_0)/R$ [17]. The second term incorporates the effect of the spin polarization induced through the interaction $U$. Fig. 4 shows that Eq. 4 provides an excellent description of the exact $\Delta p$ data as a function of $R$. The approximation continues to be equally good up to $R = 30$ Å, although for clarity results over a shorter $R$-range are shown in the figure. We emphasize that the form of our solution in Eq. 4 does not depend explicitly on that of the noninteracting Hamiltonian ($\hat{H}_0$). Although details of $\zeta$ will vary with those of the spectrum of $\hat{H}_0$, we expect our prediction of oscillations in $\zeta$ with dot radius to be generally robust to changes in the shape and dimensionality of the confining potential [20,21]. Thus, we expect our results to be relevant to almost one dimensional systems. Indeed, in metallic nanowires [22], Zabala et al. [4] report a series of peaks in the magnetic moment per electron
as a function of wire radius, correlated strongly with another observable of the system (the elongation force of the nanowire). The physics driving these results is similar to that in our work \cite{23}, despite the fact that Ref. \cite{2} considers the stabilized jellium model within the framework of the spin-dependent density-functional formalism, while we treat correlation effects differently, in terms of the direct interaction $U$ and neglect various off-diagonal exchange and Coulomb contributions \cite{24}.

The relationship between $\zeta$ and $\Delta p$ provides a powerful handle for a direct experimental verification of the polarization oscillations in quantum dots predicted in this study via the use of solid state spectroscopies sensitive to the EMD, namely, angular-correlation of positron-annihilation radiation (ACAR) \cite{18}, positrons have the advantage of being local probes, see Weber \textit{et al.} in Ref. \cite{15}, inelastic X-ray scattering (IXS) in the deeply inelastic (Compton) regime and scanning tunneling microscopy (STM). We have carried out further computations at other $r_s$ values and find that, generally speaking, polarization effects become less prominent with decreasing $r_s$, as the kinetic energy dominates, as well as with increasing $R$, as electrons are less confined and $U$ becomes weaker. Nevertheless, oscillations in $\Delta p$ are observable even for $r_s \simeq 3$, particularly for dot sizes $R \lesssim 10 \text{ Å}$.

In summary, we have presented an \textit{exactly solvable} model Hamiltonian for discussing the properties of the interacting electron gas in the confined geometry of a quantum dot. Although the results presented in this article are based on a confining potential in the shape of a spherical square well, computations for other geometries and confining potentials would be quite straightforward. The ground state is found to exhibit spontaneous magnetization and striking oscillations in spin polarization $\zeta$ as a function of the dot radius $R$, at the fixed electron density considered. The oscillations in $\zeta$ are shown to induce similar oscillations in the width $\Delta p$ around the Fermi cut-off in the momentum density, which we refer to for simplicity as the dot “Fermi momentum”, providing a novel route for direct experimental observation of the dot magnetization via spectroscopies sensitive to momentum density (specially, positron annihilation and Compton scattering). A simple expression for $\Delta p$ is discussed, which gives an excellent approximation to the exact numerical data as a function of radius. We expect the results of the present study to be robust to the details of the confining potential and to the approximations inherent in the treatment of correlations in our model Hamiltonian.

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  \bibitem{16} Note deviations from first Hund’s rule filling in 2D quantum dots have long been reported. See, e.g., Ref. \cite{18}.
  \bibitem{17} One has $\Delta p > 0$ also in bulk semiconductors and metals along directions in the Brillouin zone where the spectrum contains a band gap (e.g., along [111] in Cu). In these cases the cutoff momentum is related to the dimension of the Jones or Brillouin zones, respectively. See M. H. Weber \textit{et al.}, Phys. Rev. B \textbf{66}, 041305(R) (2002).
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We do not contradict the theorem of Lieb and Mattis [Phys. Rev. 125, 164 (1962)] regarding the absence of a magnetic ground state in the 1D case because our interaction in Eq. (1) is spin dependent.

Even if the confining potential is deformed, to observe oscillations in $\zeta$ it will suffice to have singularities in the noninteracting density of states and a strong $U$.

A nanowire can be viewed as a very elongated ellipsoid.

Our $UN$ is close to the Stoner parameter $I$ of Ref. 6, both in meaning and in value.

These off-diagonal terms are generally expected to reduce the ground state polarization.