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Electron correlations in an excited state of a quantum dot in a uniform magnetic field

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ABSTRACT

Electron correlations in a two-electron two-dimensional 'artificial atom' or quantum dot (with harmonic confining potential) in the presence of a uniform magnetic field in an excited singlet state are studied via quantal density functional theory (QDFT). QDFT allows for the separation of the electron correlations due to the Pauli exclusion principle and Coulomb repulsion, as well as the determination of the contribution of these correlations to the kinetic energy. The QDFT mapping is from the excited state of the quantum dot to one of noninteracting fermions in their ground state possessing the same basic variables of the density and physical current density, and the same orbital and spin angular momentum. A detailed analysis of these correlations in terms of their quantal sources, the corresponding 'classical' fields, and resulting potentials and energies is presented. The key conclusions are that as in natural atoms, the contributions of the Pauli and Coulomb correlations relative to the total energy for the excited state, are less than but of the same order of magnitude as those for the ground state of a quantum dot. However, in contrast, the correlation-kinetic contributions are an order of magnitude greater than those for a quantum dot in its ground state. These correlations constitute nearly 75% of the kinetic and 25% of the total energy. This result is consistent with prior work on low electron density Wigner systems in three-dimensions in which correlation-kinetic effects too play a significant role. The significance of these correlations to traditional excited state density functional theory is noted.

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1. Introduction

As a result of advances in semiconductor technology over the past few decades, it is possible to create 'artificial atoms' in which the motion of the electrons is confined to two dimensions [1–4]. This is achieved by creating a thin (10 nm) quantum well within a layer of a semiconductor (GaAs) of lateral width 100 nm sandwiched between two layers of another semiconductor (AlGaAs). There is no motion perpendicular to the well. The free motion of the electrons laterally is confined by a field so as to create the 'artificial atom' or quantum dot. The size of the quantum dot can be further reduced by application of a perpendicular magnetic field. The few electron 'artificial atom' or quantum dot possesses the same electronic structure characteristics as that of a regular atom. There is, however, a fundamental difference between the natural and 'artificial' atom which is arrived at via both experiment [5–8] and theory [9] performed on the latter. Whereas in a natural atom the electrons are confined to the nucleus by a Coulomb potential, those in the quantum dot are confined to the atom center harmonically. In particular, there is support for the harmonic confinement via the Generalized Kohn theorem [3,10–16]. As a consequence, the wave function of a quantum dot does not exhibit a cusp at the atom center. It does satisfy the two-dimensional electron-electron coalescence constraint [17]. Another important difference is that the size of the quantum dot can be about an order of magnitude greater than atoms occurring in nature: 2–6 nm vs 0.1 nm. As a result, there is a lowering of the electron density, and in a manner akin to the Wigner system [18–25], electron correlation effects become more significant. This has been confirmed by various calculations [3,4,26–29] on quantum dots. A striking result [29] obtained via quantal density functional theory [30,31] for the ground state of a two-electron quantum dot in a uniform magnetic field was the significance of correlation-kinetic effects. These are contributions to the kinetic energy that are solely due to correlations between the electrons. The contribution of correlation-kinetic effects to the total energy was determined to be greater than those of the Coulomb contributions, and over ten percent of those due to the Pauli exclusion principle. The significance of these correlation-kinetic effects thus mirror what occurs

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in the low density Wigner regime [18,19]. In the present paper we study the electron correlations in an excited singlet state of a quantum dot (with a harmonically confining potential) in the presence of a uniform magnetic field. The study is performed via quantal density functional theory (QDFT) [30,31].

The quantum dot to be studied may be described as a system of N electrons in an electrostatic $\boldsymbol{\mathcal{E}}(\mathbf{r}) = -\nabla v(\mathbf{r})$ and magnetostatic $\boldsymbol{\mathcal{B}}(\mathbf{r}) = \nabla \times \boldsymbol{\mathcal{A}}(\mathbf{r})$ field with orbital $\mathbf{L}$ and spin $\mathbf{S}$ angular momentum, and where $\{v(\mathbf{r}), \boldsymbol{\mathcal{A}}(\mathbf{r})\}$ are the scalar and vector potentials. Stationary-state QDFT in this instance [30,32] constitutes the mapping from any state of such a system to one of noninteracting fermions possessing the same density $\rho(\mathbf{r})$, physical current density $\mathbf{j}(\mathbf{r})$, orbital $\mathbf{L}$ and spin $\mathbf{S}$ angular momenta. The reason for the mapping to a model system with the same properties $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ stems from the first Hohenberg-Kohn theorem [33]. The theorem proves that in the presence of only an electrostatic field there is a bijective relationship between the nondegenerate ground state density $\rho(\mathbf{r})$ and the external scalar potential $v(\mathbf{r})$. The constraint in the proof is that of fixed electron number N. Hence, knowledge of this density uniquely determines the system Hamiltonian to within a constant, and thereby via solution of the Schrödinger equation, the wave functions of the system. The ground state density is thus said to be a basic variable of quantum mechanics.

In the added presence of a magnetic field in which the interaction of the field with both the orbital and spin angular momenta is considered, the corresponding Schrödinger-Pauli Hamiltonian [34,35] in a.u. (charge of electron $-e$, $|e| = \hbar = m = 1$ together with the assumption of $c = 1$) is

$$\widehat{H} = \frac{1}{2}\sum_k [\hat{\mathbf{p}}_k + \boldsymbol{\mathcal{A}}(\mathbf{r}_k)]^2 + \frac{1}{2}\sum_{k,l}{}' \frac{1}{|\mathbf{r}_k - \mathbf{r}_l|} + \sum_k v(\mathbf{r}_k) + \sum_k \boldsymbol{\mathcal{B}}(\mathbf{r}_k)\cdot \mathbf{s}_k, \tag{1}$$

where the operator terms correspond to the physical kinetic (with canonical momentum $\hat{\mathbf{p}}_k = -i\nabla_{\mathbf{r}_k}$), electron-interaction, external potential, and magnetic field-spin interaction with $\mathbf{s}$ the electron spin momentum vector. In recent work, it has been proved [30,35] that in the presence of an electrostatic and *uniform* magnetostatic field $\boldsymbol{\mathcal{B}}(\mathbf{r}) = B\boldsymbol{i}_z$ the basic variables are the nondegenerate ground state densities $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$. In this case the constraints are those of fixed electron number N, orbital $\mathbf{L}$ and spin $\mathbf{S}$ angular momentum. Thus, knowledge of this $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ uniquely determines the scalar $v(\mathbf{r})$ and vector $\boldsymbol{\mathcal{A}}(\mathbf{r})$ potentials to within a constant and the gradient of a scalar function, respectively, thereby the Hamiltonian, and consequently the system wave functions. When the interaction of the magnetic field is only with the orbital angular momentum, then the last term in the above Hamiltonian is absent. In this case the basic variables are again the densities $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ with the constraints of fixed electron number N and orbital angular momentum $\mathbf{L}$. This constitutes a special case.

An attribute of QDFT is that it is possible to separate the contributions to the total energy from electron correlations due to Coulomb repulsion and those arising from the Pauli exclusion principle for the *same* system. (The definition of Coulomb correlations in traditional quantum chemistry differs in that a separate Hartree-Fock theory calculation corresponding to a different density needs to be performed.) Additionally, it is possible to separately obtain the correlation contributions to the kinetic energy, *viz.* the correlation-kinetic component of the energy.

Finally, within QDFT, if in addition to possessing the same densities $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$, the model fermions are subject to the same external potentials $\{v(\mathbf{r}), \boldsymbol{\mathcal{A}}(\mathbf{r})\}$, then it is proved [30,32] that the only correlations that must be accounted for in the mapping are those due to the Pauli exclusion principle, Coulomb repulsion and correlation-kinetic effects. Thus, it is only these correlations that appear in the expressions for the electron-interaction component of the local effective potential and total energy of the model fermions.

The equations of QDFT in the presence of a uniform magnetic field incorporating the recent developments [30,32,35] are given in Section 2. The application of QDFT to a quantum dot in an excited singlet state together with a discussion of the results is provided in Section 3. The principal conclusions are summarized in Section 4. The relevance of the present results to traditional excited state density functional theory is also discussed in this section.

2. Quantal density functional theory

For a system of N electrons in an external electrostatic $\boldsymbol{\mathcal{E}}(\mathbf{r}) = -\nabla v(\mathbf{r})$ and magnetostatic $\boldsymbol{\mathcal{B}}(\mathbf{r}) = \nabla \times \boldsymbol{\mathcal{A}}(\mathbf{r})$ field, and in a singlet state, the Hamiltonian of Eq. (1) reduces to

$$\widehat{H} = \frac{1}{2}\sum_k [\hat{\mathbf{p}}_k + \boldsymbol{\mathcal{A}}(\mathbf{r}_k)]^2 + \frac{1}{2}\sum_{k,l}{}' \frac{1}{|\mathbf{r}_k - \mathbf{r}_l|} + \sum_k v(\mathbf{r}_k), \tag{2}$$

with the corresponding Schrödinger equation being

$$\widehat{H}\Psi(\mathbf{X}) = E\Psi(\mathbf{X}), \tag{3}$$

$\{\Psi(\mathbf{X}), E\}$ the eigenfunctions and eigenvalues; $\mathbf{X} = \mathbf{x}_1, \ldots, \mathbf{x}_N$; $\mathbf{x} = \mathbf{r}\sigma$; $(\mathbf{r}\sigma)$ the spatial and spin coordinates of each electron. The energy E is the expectation $E = \langle\Psi(\mathbf{X})|\widehat{H}|\Psi(\mathbf{X})\rangle$. The corresponding density $\rho(\mathbf{r})$ and physical current density $\mathbf{j}(\mathbf{r})$ are the expectations

$$\rho(\mathbf{r}) = \langle\Psi(\mathbf{X})|\hat{\rho}(\mathbf{r})|\Psi(\mathbf{X})\rangle, \tag{4}$$

and

$$\mathbf{j}(\mathbf{r}) = \langle\Psi(\mathbf{X})|\hat{\mathbf{j}}(\mathbf{r})|\Psi(\mathbf{X})\rangle, \tag{5}$$

where the respective operators are

$$\rho(\mathbf{r}) = \sum_k \delta(\mathbf{r} - \mathbf{r}_k), \tag{6}$$

and

$$\hat{\mathbf{j}}(\mathbf{r}) = \frac{1}{2i}\sum_k \left[\nabla_{\mathbf{r}_k}\delta(\mathbf{r}_k - \mathbf{r}) + \delta(\mathbf{r}_k - \mathbf{r})\nabla_{\mathbf{r}_k}\right] + \hat{\rho}(\mathbf{r})\boldsymbol{\mathcal{A}}(\mathbf{r}). \tag{7}$$

The system of electrons defined by the Hamiltonian of Eq. (2) is then mapped via QDFT to one of noninteracting fermions possessing the same $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ and subject to the same external fields. The corresponding differential equation for the model fermion orbitals $\phi_k(\mathbf{x})$ is

$$\left[\frac{1}{2}(\hat{\mathbf{p}}_k + \boldsymbol{\mathcal{A}}(\mathbf{r}_k))^2 + v_s(\mathbf{r})\right]\phi_k(\mathbf{x}) = \epsilon_k\phi_k(\mathbf{x}): \quad k = 1, \ldots, N, \tag{8}$$

with the local potential

$$v_s(\mathbf{r}) = v(\mathbf{r}) + v_{ee}(\mathbf{r}), \tag{9}$$

and where $v_{ee}(\mathbf{r})$ is the local electron-interaction potential in which all the many-body effects are incorporated. The corresponding wave function is the Slater determinant $\Phi\{\phi_k\}$ with the $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ being the expectations of the operators $\{\hat{\rho}(\mathbf{r}), \hat{\mathbf{j}}(\mathbf{r})\}$ taken with respect to $\Phi\{\phi_k\}$.

The potential $v_{ee}(\mathbf{r})$ is the work done to move a model fermion from a reference point at infinity to its position at $\mathbf{r}$ in the force of a conservative effective field $\boldsymbol{\mathcal{F}}^{\text{eff}}(\mathbf{r})$:

$$v_{ee}(\mathbf{r}) = -\int_\infty^{\mathbf{r}} \boldsymbol{\mathcal{F}}^{\text{eff}}(\mathbf{r}')\cdot d\boldsymbol{\ell}', \tag{10}$$

where

$$\boldsymbol{\mathcal{F}}^{\text{eff}}(\mathbf{r}) = \boldsymbol{\mathcal{E}}_{ee}(\mathbf{r}) + \boldsymbol{\mathcal{Z}}_{t_c}(\mathbf{r}), \tag{11}$$

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