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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.

Keywords:

Quantum dot

Electron correlations

I. 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 ($10nm$) quantum well within a layer of a semiconductor (GaAs) of lateral width $100nm$ 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: $2 - 6nm$ vs $0.1nm$. 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 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 $\mathcal{E}(\mathbf{r}) = -\nabla v(\mathbf{r})$ and magnetostatic $\mathcal{B}(\mathbf{r}) = \nabla \times \mathcal{A}(\mathbf{r})$ field with orbital $\mathbf{L}$ and spin $\mathbf{S}$ angular momentum, and where $\{v(\mathbf{r}), \mathcal{A}(\mathbf{r})\}$ 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

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

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 $\mathcal{B}(\mathbf{r}) = B\mathbf{i}_z$ 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 $\mathcal{A}(\mathbf{r})$ 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 $\{v(\mathbf{r}), \mathcal{A}(\mathbf{r})\}$, 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 Sect. II. The application of QDFT to a quantum dot in an excited singlet state together with a discussion of the results is provided in Sect. III. The principal conclusions are summarized in Sect. IV. The relevance of the present results to traditional excited state density functional theory is also discussed in this section.

II. QUANTAL DENSITY FUNCTIONAL THEORY

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

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

with the corresponding Schrödinger equation being

$$\hat{H}\Psi(\mathbf{X}) = E\Psi(\mathbf{X}), \quad (3)$$

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

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

and

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

where the respective operators are

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

and

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

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

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

with the local potential

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

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 $\mathcal{F}^{\text{eff}}(\mathbf{r})$:

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

where

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

is the sum of the electron-interaction $\mathcal{E}_{ee}(\mathbf{r})$ and correlation-kinetic $\mathcal{Z}_{tc}(\mathbf{r})$ fields. As $\nabla \times \mathcal{F}^{\text{eff}}(\mathbf{r}) = 0$, the work done $v_{ee}(\mathbf{r})$ is path-independent.

The electron-interaction field $\mathcal{E}_{ee}(\mathbf{r})$ is representative of electron correlations due to the Pauli exclusion principle and Coulomb repulsion. It is defined in terms of the electron-interaction ‘force’ $\mathbf{e}_{ee}(\mathbf{r})$ and (charge) density $\rho(\mathbf{r})$ as

$$\mathcal{E}_{ee}(\mathbf{r}) = \frac{\mathbf{e}_{ee}(\mathbf{r})}{\rho(\mathbf{r})}, \quad (12)$$

where $\mathbf{e}_{ee}(\mathbf{r})$ is obtained via Coulomb's law from its quantal source, the pair-correlation function $P(\mathbf{r}\mathbf{r}')$:

$$\mathbf{e}_{ee}(\mathbf{r}) = \int \frac{P(\mathbf{r}\mathbf{r}')(\mathbf{r} - \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|^3} d\mathbf{r}', \quad (13)$$

with $P(\mathbf{r}\mathbf{r}')$ the expectation

$$P(\mathbf{r}\mathbf{r}') = \langle \Psi(\mathbf{X}) | \hat{P}(\mathbf{r}\mathbf{r}') | \Psi(\mathbf{X}) \rangle \quad (14)$$

of the pair-operator

$$\hat{P}(\mathbf{r}\mathbf{r}') = \sum'_{k,l} \delta(\mathbf{r}_k - \mathbf{r}) \delta(\mathbf{r}_l - \mathbf{r}'). \quad (15)$$

The correlation-kinetic field $\mathbf{Z}_{t_c}(\mathbf{r})$ is the difference between the kinetic fields $\mathbf{Z}(\mathbf{r})$, $\mathbf{Z}_s(\mathbf{r})$ of the interacting and model fermion systems, respectively. These fields are defined in terms of the respective kinetic 'forces' $\mathbf{z}(\mathbf{r})$, $\mathbf{z}_s(\mathbf{r})$ and the density $\rho(\mathbf{r})$. Thus,

$$\mathbf{Z}_{t_c}(\mathbf{r}) = \mathbf{Z}_s(\mathbf{r}) - \mathbf{Z}(\mathbf{r}) = \frac{\mathbf{z}_s(\mathbf{r}) - \mathbf{z}(\mathbf{r})}{\rho(\mathbf{r})}. \quad (16)$$

The kinetic 'force' $\mathbf{z}(\mathbf{r})$ is obtained from its quantal source, the single-particle density matrix $\gamma(\mathbf{r}\mathbf{r}')$:

$$z_\alpha(\mathbf{r}) = 2 \sum_\beta \nabla_\beta t_{\alpha\beta}(\mathbf{r}), \quad (17)$$

where the kinetic energy tensor is

$$t_{\alpha\beta}(\mathbf{r}) = \frac{1}{4} \left[\frac{\partial^2}{\partial r'_\alpha \partial r''_\beta} + \frac{\partial^2}{\partial r'_\beta \partial r''_\alpha} \right] \gamma(\mathbf{r}'\mathbf{r}'') \Big|_{\mathbf{r}'=\mathbf{r}''=\mathbf{r}}, \quad (18)$$

with $\gamma(\mathbf{r}\mathbf{r}')$ the expectation

$$\gamma(\mathbf{r}\mathbf{r}') = \langle \Psi(\mathbf{X}) | \hat{\gamma}(\mathbf{r}\mathbf{r}') | \Psi(\mathbf{X}) \rangle \quad (19)$$

of the density matrix operator

$$\hat{\gamma}(\mathbf{r}\mathbf{r}') = \hat{A} + i\hat{B}, \quad (20)$$

$$\hat{A} = \frac{1}{2} \sum_j [\delta(\mathbf{r}_j - \mathbf{r}) T_j(\mathbf{a}) + \delta(\mathbf{r}_j - \mathbf{r}') T_j(-\mathbf{a})], \quad (21)$$

$$\hat{B} = \frac{i}{2} \sum_j [\delta(\mathbf{r}_j - \mathbf{r}) T_j(\mathbf{a}) + \delta(\mathbf{r}_j - \mathbf{r}') T_j(-\mathbf{a})], \quad (22)$$

and $T_j(\mathbf{a})$ a translation operator such that $T_j(\mathbf{a})\psi(\dots\mathbf{r}_j\dots) = \psi(\dots\mathbf{r}_j + \mathbf{a},\dots)$. The definition of the 'force' $\mathbf{z}_s(\mathbf{r})$ is the same except that the quantal source is the Dirac density matrix $\gamma_s(\mathbf{r}\mathbf{r}')$ which is the expectation

$$\gamma_s(\mathbf{r}\mathbf{r}') = \langle \Phi\{\phi_k\} | \hat{\gamma}(\mathbf{r}\mathbf{r}') | \Phi\{\phi_k\} \rangle. \quad (23)$$

The total energy E as obtained from the model system may be written in terms of the individual fields as

$$E = \sum_k \epsilon_k - \int \rho(\mathbf{r}) v_{ee}(\mathbf{r}) + E_{ee} + T_c, \quad (24)$$

or as

$$E = T_s + \int \rho(\mathbf{r}) v(\mathbf{r}) d\mathbf{r} + E_{ee} + T_c, \quad (25)$$

where the model system kinetic energy is

$$T_s = -\frac{1}{2} \int \rho(\mathbf{r})(\mathbf{r}) \cdot \mathbf{Z}_s(\mathbf{r}) d\mathbf{r}, \quad (26)$$

the electron-interaction energy is

$$E_{ee} = \int \rho(\mathbf{r})(\mathbf{r}) \cdot \mathbf{E}_{ee}(\mathbf{r}) d\mathbf{r}, \quad (27)$$

and the correlation-kinetic energy is

$$T_c = \frac{1}{2} \int \rho(\mathbf{r})(\mathbf{r}) \cdot \mathbf{Z}_{tc}(\mathbf{r}) d\mathbf{r}. \quad (28)$$

Finally, it is possible to subdivide the electron-interaction field $\mathbf{E}_{ee}(\mathbf{r})$, and therefore the corresponding energy component E_{ee} , into its Hartree $[\mathbf{E}_H(\mathbf{r}), E_H]$, Pauli $[\mathbf{E}_x(\mathbf{r}), E_x]$, and Coulomb $[\mathbf{E}_c(\mathbf{r}), E_c]$ components. The pair-correlation density for the interacting system is $g(\mathbf{r}\mathbf{r}') = P(\mathbf{r}\mathbf{r}')/\rho(\mathbf{r}) = \rho(\mathbf{r}') + \rho_{xc}(\mathbf{r}\mathbf{r}')$, where $\rho_{xc}(\mathbf{r}\mathbf{r}')$ is the Fermi-Coulomb hole charge. The pair-correlation density for the model system is $g_s(\mathbf{r}\mathbf{r}') = P_s(\mathbf{r}\mathbf{r}')/\rho(\mathbf{r}) = \rho(\mathbf{r}') + \rho_x(\mathbf{r}\mathbf{r}')$, where $P_s(\mathbf{r}\mathbf{r}') = \langle \Phi\{\phi_k\} | \hat{P}(\mathbf{r}\mathbf{r}') | \Phi\{\phi_k\} \rangle$, and $\rho_x(\mathbf{r}\mathbf{r}') = -|\gamma_s(\mathbf{r}\mathbf{r}')|^2/2\rho(\mathbf{r})$ is the Fermi hole charge. The Coulomb hole charge is defined as $\rho_c(\mathbf{r}\mathbf{r}') = \rho_{xc}(\mathbf{r}\mathbf{r}') - \rho_x(\mathbf{r}\mathbf{r}')$. The expressions for the individual component fields and energies are the same as those for $[\mathbf{E}_{ee}(\mathbf{r}), E_{ee}]$ but with quantal sources $\rho(\mathbf{r}), \rho_x(\mathbf{r}\mathbf{r}')$, and $\rho_c(\mathbf{r}\mathbf{r}')$. Provided the fields $\mathbf{E}_x(\mathbf{r}), \mathbf{E}_c(\mathbf{r}), \mathbf{Z}_{tc}(\mathbf{r})$ are conservative, then the electron-interaction potential $v_{ee}(\mathbf{r})$ may also be subdivided into its electron-interaction $W_{ee}(\mathbf{r})$ and correlation-kinetic components. The potential $W_{ee}(\mathbf{r})$ may be further subdivided into its Hartree $W_H(\mathbf{r})$, Pauli $W_x(\mathbf{r})$, and Coulomb $W_c(\mathbf{r})$ components:

$$v_{ee}(\mathbf{r}) = W_{ee}(\mathbf{r}) + W_{tc}(\mathbf{r}) = W_H(\mathbf{r}) + W_x(\mathbf{r}) + W_c(\mathbf{r}) + W_{tc}(\mathbf{r}). \quad (29)$$

Each potential component is the work done in the corresponding field.

III. APPLICATION TO A QUANTUM DOT IN AN EXCITED SINGLET STATE

The Hamiltonian of a two-electron quantum dot in a magnetic field $\mathcal{B}(\mathbf{r}) = \nabla \times \mathcal{A}(\mathbf{r})$ and in a singlet state is

$$\hat{H} = \sum_{k=1}^2 \left\{ \frac{1}{2} (\hat{\mathbf{p}}_k + \mathcal{A}(\mathbf{r}_k))^2 + \frac{1}{2} \omega_0^2 r_k^2 \right\} + \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|}. \quad (30)$$

In the symmetric gauge $\mathcal{A}(\mathbf{r}) = \frac{1}{2} \mathcal{B}(\mathbf{r}) \times \mathbf{r}$ with the magnetic field applied in the z -direction: $\mathcal{B}(\mathbf{r}) = B \hat{\mathbf{i}}_z$, the first excited singlet state 2^1S wave function following Taut [36–40] is derived to be

$$\Psi_{01}(\mathbf{r}_1 \mathbf{r}_2) = \Psi_{01}(\mathbf{R} \mathbf{r}) = \xi_0(\mathbf{R}) \phi_1(\mathbf{r}), \quad (31)$$

with $\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$ the center of mass, and $\mathbf{r} = \mathbf{r}_2 - \mathbf{r}_1$ the relative coordinates. Defining an effective force constant $k_{\text{eff}} = \Omega^2 = \omega_0^2 + \omega_L^2$, where $\omega_L = B/2$ is the Larmor frequency, the center of mass component of the wave function is

$$\xi_0(\mathbf{R}) = C_{0R} e^{-\sqrt{k_{\text{eff}}} R^2}, \quad (32)$$

where $C_{0R} = \sqrt{2\Omega/\pi}$. For the 2^1S state $k_{\text{eff}} = 0.471716$, $\Omega = 0.686816$, and $C_{0R} = 0.661242$. The relative coordinate component of the wave function is

$$\phi_1(\mathbf{r}) = C_{1r} e^{-\frac{1}{4}\sqrt{k_{\text{eff}}} r^2} (1 + r + ar^2 + br^3), \quad (33)$$

with $C_{1r} = 0.164181$, $a = -0.265111$, $b = -0.182082$. The energy of this state is $E_1 = 3.434066$ *a.u.* (See Table I)

Fig. 1 is a plot of the relative coordinate function $\phi_1(\mathbf{r})$. Observe the node for this excited state. Also observe the cusp [8] for electron-electron coalescence. The wave function $\Psi_{01}(\mathbf{r}_1 \mathbf{r}_2)$ is plotted in Figs. 2-4 for different angles $\theta' = 0^\circ, 45^\circ, 90^\circ$ between the vectors $\mathbf{r}_1$ and $\mathbf{r}_2$, and for different orientations of these vectors. In each figure the node in the wave function is clearly evident. Observe the electron-electron cusp as exhibited by the wave function for $\mathbf{r}_1 = \mathbf{r}_2$ in Fig. 2.

The above interacting system in the 2^1S state is next mapped to one of noninteracting fermions in their ground state with the same density $\rho_1(\mathbf{r})$, physical current density $\mathbf{j}_1(\mathbf{r})$, orbital $\mathbf{L}$ and spin $\mathbf{S}$ angular momenta. The corresponding Schrödinger equation for the model system is then

$$\left[\frac{1}{2} \hat{p}^2 + \frac{1}{2} k_{\text{eff}} r^2 + v_{\text{ee}}(\mathbf{r}) \right] \phi_k(\mathbf{r}) = \epsilon_k \phi_k(\mathbf{r}), \quad k = 1, 2 \quad (34)$$

with

$$\phi_k(\mathbf{r}) = \sqrt{\rho_1(\mathbf{r})/2}, \quad (35)$$

and where $v_{ee}(\mathbf{r})$ is defined by Eqs. (10), (11). Many properties of this mapping can be obtained in closed analytical or semianalytical form. The asymptotic structure of these properties in the classically forbidden region and near the center of the quantum dot can also be obtained analytically. These expressions are given in Appendix A. We next discuss the various properties.

A. Quantal sources

1. Electron density $\rho_1(\mathbf{r})$ and physical current density $\mathbf{j}_1(\mathbf{r})$

The analytical expression for the density $\rho_1(\mathbf{r})$ and its asymptotic structure is given in Appendix A. The electron density $\rho_1(\mathbf{r})$ (which has cylindrical symmetry: $\rho_1(\mathbf{r}) = \rho_1(r)$), and the radial probability density $r\rho_1(r)$, are plotted in Fig. 5. Observe that the electron density does not exhibit a cusp at the center of the quantum dot but approaches it quadratically. The ‘artificial atom’ exhibits shell structure. This is clearly evident in the plots for both the density $\rho_1(r)$ as well as the radial probability density $r\rho_1(r)$. As the wave function is real, the physical current density $\mathbf{j}_1(\mathbf{r}) = \rho_1(\mathbf{r})\mathbf{A}(\mathbf{r})$. It satisfies the continuity condition $\nabla \cdot \mathbf{j}_1(\mathbf{r}) = 0$.

2. Pair-correlation density $g(\mathbf{r}\mathbf{r}')$, Single-particle $\gamma(\mathbf{r}\mathbf{r}')$ and Dirac $\gamma_s(\mathbf{r}\mathbf{r}')$ density matrices

The expressions for the quantal sources, the pair-correlation density $g(\mathbf{r}\mathbf{r}')$, the density matrix $\gamma(\mathbf{r}\mathbf{r}')$, and the Dirac density matrix $\gamma_s(\mathbf{r}\mathbf{r}')$ are given in Appendix A. From these expressions follow those for the quantal sources of the Fermi $\rho_x(\mathbf{r}\mathbf{r}')$ and Coulomb $\rho_c(\mathbf{r}\mathbf{r}')$ holes. For the present mapping, the Fermi hole $\rho_x(\mathbf{r}\mathbf{r}') = -\rho_1(\mathbf{r}')/2$ independent of the electron position at $\mathbf{r}$. Thus, the nonlocality of the pair-correlation density $g(\mathbf{r}\mathbf{r}')$ is exhibited either via the Fermi-Coulomb hole $\rho_{xc}(\mathbf{r}\mathbf{r}')$ or the Coulomb hole $\rho_c(\mathbf{r}\mathbf{r}')$.

In Fig. 6a, the cross-sections through the Fermi-Coulomb $\rho_{xc}(\mathbf{r}\mathbf{r}')$, Fermi $\rho_x(\mathbf{r}\mathbf{r}')$, and Coulomb $\rho_c(\mathbf{r}\mathbf{r}')$ holes for an electron at the center of the quantum dot at $r = 0$ are plotted. The electron position is indicated by the arrow. Observe that for this electron position, these charge distributions are all cylindrically symmetric about the z -axis. (As a consequence, the corresponding fields vanish at this point.) Also observe how the electron-electron coalescence condition on the wave function [17, 31] is exhibited in the corresponding Fermi-Coulomb and Coulomb hole distributions. A better pictorial sense of the full structure of the Coulomb

hole $\rho_c(\mathbf{r}\mathbf{r}')$ is obtained via Fig. 6b in which a surface plot of the hole for the same electron position is presented. Here too, observe the electron-electron coalescence cusp at the electron position. (For this plot, x' is the projection of $\mathbf{r}'$ on $\mathbf{r}$, *i.e.* $x' = r'\hat{\mathbf{r}} \cdot \hat{\mathbf{r}}'$, and y' is the projection of $\mathbf{r}'$ on the direction perpendicular to $\mathbf{r}$, *i.e.* $y' = r'[1 - (\hat{\mathbf{r}} \cdot \hat{\mathbf{r}}')^2]^{\frac{1}{2}}$.)

In the panels (a) of Figs. 7 - 10, cross sections of the Coulomb hole $\rho_c(\mathbf{r}\mathbf{r}')$ in different directions $\theta' = 0^\circ, 45^\circ, 90^\circ$ with respect to the center of quantum dot - electron direction are plotted. The electron positions considered are at $r = 0.5, 1, 2, 20$ *a.u.*. In the panels (b), the surface plots of the Coulomb hole $\rho_c(\mathbf{r}\mathbf{r}')$ for the same electron positions are shown. Observe the nonlocal (dynamic) structure of the Coulomb hole, and the fact that it is not symmetrical about the electron position. In Figs. 7, 8, the electron-electron coalescence cusp is also clearly exhibited. The cusp is too weak to be exhibited in Figs. 9, 10 on the scale of these figures. Note that for asymptotic positions of the electron (Fig. 10), the Coulomb hole is becoming essentially cylindrically symmetric and static. For far asymptotic positions of the electron in the classically forbidden region, the hole is symmetric and static.

B. Fields, Potentials and Energies

1. Fields

The analytical expressions for the electron-interaction field $\mathcal{E}_{ee}(\mathbf{r})$ and of its asymptotic structure in the classically forbidden region and near the center of the quantum dot is given in Appendix A. It's Hartree $\mathcal{E}_H(\mathbf{r})$, Pauli $\mathcal{E}_x(\mathbf{r})$, and Coulomb $\mathcal{E}_c(\mathbf{r})$ components are plotted in Fig. 11. Note that since the quantal sources of the density $\rho_1(\mathbf{r})$, Fermi $\rho_x(\mathbf{r}\mathbf{r}')$, and Coulomb $\rho_c(\mathbf{r}\mathbf{r}')$ holes are cylindrically symmetric for an electron at the center, the respective fields vanish there. In proceeding away from the center, each field is seen to exhibit shell structure. Finally, as the density $\rho_1(\mathbf{r})$ and the Fermi $\rho_x(\mathbf{r}\mathbf{r}')$ hole are static charge distributions satisfying the constraints $\int \rho_1(\mathbf{r})d\mathbf{r} = 2$ and $\int \rho_x(\mathbf{r}\mathbf{r}')d\mathbf{r}' = -1$, the asymptotic structure of the Hartree and Pauli fields are, respectively, $\mathcal{E}_H(\mathbf{r}) \sim 2/r^2$ and $\mathcal{E}_x(\mathbf{r}) \sim -1/r^2$. The Coulomb field decays as $\mathcal{E}_c(\mathbf{r}) \sim O(-10.1/r^4)$.

The expressions for the kinetic energy tensor for the interacting $t_{\alpha\beta}(\mathbf{r}; \gamma)$ and noninteracting $t_{s,\alpha\beta}(\mathbf{r}; \gamma_s)$ systems, and those for the corresponding forces $z_\alpha(\mathbf{r}; \gamma)$ and $z_{s,\alpha}(\mathbf{r}; \gamma_s)$, as well as the asymptotic structures of the latter are given in Appendix A. The derivation of the interacting system kinetic energy tensor $t_{\alpha\beta}(\mathbf{r}; \gamma)$ and for the corresponding kinetic 'force' $z_\alpha(\mathbf{r}, \gamma)$ are given in Appendix B. The kinetic 'forces' $z(r)$ and $z_s(r)$ are plotted in Fig. 12a. The correlation-kinetic field $\mathcal{Z}_{t_c}(\mathbf{r})$ is given in Fig. 12b. This field too vanishes

at the origin, exhibits shell structure, and decays asymptotically as $O(9/r^3)$.

A comparison of all the fields is made in Fig. 13. Observe that the magnitude of the correlation-kinetic field $\mathcal{Z}_{t_c}(\mathbf{r})$ is much larger than those of the components of the electron-interaction field $\mathcal{E}_{ee}(\mathbf{r})$. This will then reflect on the respective magnitudes of the energies as shown and discussed below in Table I.

2. Potentials

As a consequence of cylindrical symmetry, the electron-interaction field $\mathcal{E}_{ee}(\mathbf{r})$ and its Hartree $\mathcal{E}_H(\mathbf{r})$, Pauli $\mathcal{E}_x(\mathbf{r})$, and Coulomb $\mathcal{E}_c(\mathbf{r})$ components, and the correlation-kinetic field $\mathcal{Z}_{t_c}(\mathbf{r})$ are all conservative. Hence, the local electron-interaction potential $v_{ee}(\mathbf{r})$ of Eq. (29) can be expressed as the sum of its Hartree $W_H(\mathbf{r})$, Pauli $W_x(\mathbf{r})$, Coulomb $W_c(\mathbf{r})$, and correlation-kinetic $W_{t_c}(\mathbf{r})$ components. Each of these potential components is the work done in the corresponding field. A comparison of these potentials is given in Fig. 14. Their structure follows from that of the fields. For example, since the Pauli field $\mathcal{E}_x(\mathbf{r})$ is negative, so is the potential $W_x(\mathbf{r})$. As $\mathcal{E}_x(\mathbf{r})$ decays asymptotically as $-1/r^2$, the potential $W_x(\mathbf{r})$ decays there as $-1/r$. The Coulomb field $\mathcal{E}_c(\mathbf{r})$ is both positive and negative, and thus so is the potential $W_c(\mathbf{r})$ although it is principally negative, and so on. The asymptotic structure of the various potentials near the atom center and the classically forbidden region is given in Appendix A. In particular, we note that even though the correlation-kinetic field $\mathcal{Z}_{t_c}(\mathbf{r})$ is both positive and negative (indicative of shell structure), the correlation-kinetic potential $W_{t_c}(\mathbf{r})$ is positive throughout space. (This was also the case for the ground state [29, 30] of the quantum dot.) The potential $v_{ee}(\mathbf{r})$ and its electron-interaction $W_{ee}(\mathbf{r})$ and correlation-kinetic $W_{t_c}(\mathbf{r})$ components are plotted in Fig. 15.

3. Energies

The QDFT determined properties of the model system in its ground state that reproduces the densities $\{\rho_1(\mathbf{r}), \mathbf{j}_1(\mathbf{r})\}$ and energy E of the singlet 2^1S state of the quantum dot are given in Table I. The expectations of the single-particle operators $\hat{O} = \delta(\mathbf{r}), r, r^2, 1/r$, are also quoted. Analytical expressions for various energy components and the expectations are provided in Appendix A. For purposes of comparison, we have also included in Table I the corresponding QDFT properties for the ground state of the quantum dot [14, 15].

As expected, the Pauli E_x and Coulomb E_c energies as a fraction of the total energy E for the excited state of the quantum dot are less than those for its ground state but of the same order of magnitude: 20% and 2.1% , respectively, as opposed to 30% and 2.6% (See

Table I). A key observation of the QDFT mapping from the ground state of the quantum dot [29, 30] was that due to the reduction in dimensionality to 2D, and the lowering of the density due to the size of the dot, correlation-kinetic effects play a more significant role than in 3D. Thus, in that case $T_c/T = 12\%$ and $T_c/E = 3.5\%$ (See Table I). For the quantum dot in the excited 2^1S state, the corresponding ratios are $T_c/T = 73\%$ and $T_c/E = 27\%$. Hence, correlation-kinetic effects are once again important, and in fact constitute an order of magnitude greater fraction of the total energy. This increase in the significance of the correlation-kinetic effects is foreseen in the structure of the correlation-kinetic field $\mathbf{Z}_{tc}(\mathbf{r})$. The result is also consistent with the fact that the size $\langle r \rangle$ of the quantum dot in the excited state is greater (See Table I), the overall density therefore lower (See Table I for $\langle \delta(\mathbf{r}) \rangle$), and consequently correlation-kinetic effects are more significant.

IV. CONCLUSIONS

In the present work we have applied QDFT to study electron correlations in a two-electron quantum dot or ‘artificial atom’ in the presence of a perpendicular uniform magnetic field in the first excited singlet state. The QDFT mapping is from the excited state of the quantum dot to one of noninteracting fermions in their ground state but possessing the same basic variables of the density $\rho_1(\mathbf{r})$ and the physical current density $\mathbf{j}_1(\mathbf{r})$, and the orbital $\mathbf{L}$ and spin $\mathbf{S}$ angular momentum. As a consequence of the mapping, it is possible to study the separate electron correlations due to the Pauli exclusion principle and Coulomb repulsion, and those due to the correlation contributions to the kinetic energy. A detailed analysis of these correlations - the quantal sources, the corresponding fields, and from these fields the resulting potentials and energies – and how they contribute to the properties of the quantum dot in its excited state is presented. The key conclusions are the following. The Hartree, Pauli, and Coulomb correlation contribution trends for this two-dimensional atom in its first excited singlet state are similar to those of a natural atom in such an excited state. In other words, their contributions are less than those of a quantum dot in its ground state, but of the same order of magnitude. What is strikingly different, is that the correlation-kinetic contributions are an order of magnitude greater than those of the quantum dot in its ground state. For the excited state, correlation-kinetic effects constitute 73% of the total kinetic energy, and 27% of the total energy. The corresponding percentages for the ground state are

TABLE I: Quantal density functional theory (QDFT) properties (in atomic units) of the model system in its ground state that reproduces the density, physical current density, orbital and spin angular momentum, and total energy E of the quantum dot in a magnetic field in its 2^1S state with effective force constant $k_{\text{eff}} = 0.471716$ (Column 2). Expectation values of single-particle operators are also quoted. For comparison, the corresponding properties [29, 30] for the QDFT mapping from a ground state of the quantum dot in a magnetic field with effective force constant $k_{\text{eff}} = 1$ to a model system in its ground state are shown (Column 3).

Property	Value in (a.u.)	
	QDFT mapping from	
	$2^1S \rightarrow 1^1S$ $k_{\text{eff}} = 0.471716$	$1^1S \rightarrow 1^1S$ $k_{\text{eff}} = 1$
E	3.434066	3.000000
E_{ext}	1.566907	1.295400
E_{ee}	0.600476	0.818401
E_H	1.343218	1.789832
E_x	-0.671609	-0.894916
	$\frac{ E_x }{E} = 20\%$	$\frac{ E_x }{E} = 30\%$
E_c	-0.071133	-0.076515
	$\frac{ E_c }{E} = 2.1\%$	$\frac{ E_c }{E} = 2.6\%$
T	1.266683	0.886199
T_s	0.338856	0.780987
T_c	0.927827	0.105212
	$\frac{T_c}{T} = 73\%$ $\frac{T_c}{E} = 27\%$	$\frac{T_c}{T} = 12\%$ $\frac{T_c}{E} = 3.5\%$
ϵ	2.746374	2.000000
	$\frac{\epsilon}{E} = 80\%$	$\frac{\epsilon}{E} = 67\%$
$\langle \delta(\mathbf{r}) \rangle = \rho(0)$	0.207299	0.436132
$\langle r \rangle$	3.276767	2.037894
$\langle r^2 \rangle$	6.643441	2.590800
$\langle 1/r \rangle$	1.930010	2.996873

12% and 3.5%, respectively. The significance of the correlation-kinetic effects is consistent with our prior conclusion [18, 19] that as the density is lowered these effects become more prominent. (A study of the Wigner regime of a quantum dot would undoubtedly show these effects to be further enhanced.) Additionally, for the excited state, the magnitude of the correlation-kinetic energy is greater than that of the Pauli and Coulomb energies. This result is a consequence of the fact that the magnitude of the correlation-kinetic field is greater than those of the Pauli and Coulomb fields, the energies being obtained from the fields via integral virial relationships.

In studying the ‘artificial atom’ in its excited singlet state, other characteristics of atomic structure are also observed. For example, the ‘atom’ exhibits shell structure which is observed not only via the radial probability density, but also via the density as well as the Hartree, Pauli, Coulomb and correlation-kinetic fields. The electron-electron coalescence constraints on the wave function for this two-dimensional atom are clearly exhibited by cusps in the structure of the Coulomb hole charge distribution. As expected, the structure of the Coulomb hole charge is dynamic and changes as a function of the electron position.

We conclude by noting that the results obtained for the excited state as well as those of our prior work [29] on the ground state are of relevance to traditional ground [33, 41] and excited state [42] density functional theory. The Gunnarsson-Lundqvist theorem [43, 44] extends the ground state Hohenberg-Kohn theorems to the lowest excited state with a symmetry different from that of the ground state. Hence, in such cases the energy is a functional of the corresponding excited state density. For other excited states, there is no corresponding Hohenberg-Kohn theorem. One approach [42] to such excited states requires a *bidensity* energy functional of the exact ground state density and the excited state density. For three-dimensional ground state systems, correlation-kinetic effects are usually small, and in the construction of approximate ‘exchange-correlation’ energy functionals of the density, these effects can be ignored. Thus, only correlations due to the Pauli exclusion principle and Coulomb repulsion need be considered. However, with a reduction in dimensionality and density, the correlation-kinetic effects become quite significant as shown in [18, 19, 29] and the present work. Hence, in developing approximate density and bidensity ‘exchange-correlation’ energy functionals, these effects must be accounted for.

FIG. 1: Relative coordinate component $\phi_1(\mathbf{r})$ of the 2^1S state of the quantum dot in a magnetic field.

FIG. 2: Structure of the 2^1S state wave function $\Psi_{01}(\mathbf{r}_1\mathbf{r}_2)$ of the quantum dot in a magnetic field. The angle between the vector $\mathbf{r}_1$ and $\mathbf{r}_2$ is $\Theta' = 0^\circ$. The vectors $\mathbf{r}_1$ and $\mathbf{r}_2$ are oriented along the positive and negative x -axis.

FIG. 3: The same as in Fig. 2 but for $\Theta' = 45^\circ$. Vector $\mathbf{r}_1$ is along the $\pm x$ -axis, and vector $\mathbf{r}_2$ is at 45° from the $\pm x$ -axis.

FIG. 4: The same as Fig. 1 but for $\Theta' = 90^\circ$. Vector $\mathbf{r}_1$ is along the $\pm x$ -axis, and vector $\mathbf{r}_2$ is along the $\pm y$ -axis.

FIG. 5: The electron density $\rho_1(r)$ and the radial probability density $r\rho_1(r)$.

FIG. 6: (a): Cross-sections through the Fermi-Coulomb $\rho_{xc}(\mathbf{r}\mathbf{r}')$, Fermi $\rho_x(\mathbf{r}\mathbf{r}')$, and Coulomb $\rho_c(\mathbf{r}\mathbf{r}')$ holes for an electron at the center of the quantum dot at $r = 0$. The electron position is indicated by the arrow. (b): Surface plot of the Coulomb hole $\rho_c(\mathbf{r}\mathbf{r}')$ for an electron at the same position. Here x' is the projection of $\mathbf{r}'$ on $\mathbf{r}$, *i.e.* $x' = r'\hat{\mathbf{r}} \cdot \hat{\mathbf{r}}'$, and y' is the projection of $\mathbf{r}'$ on the direction perpendicular to $\mathbf{r}$, *i.e.* $y' = r'[1 - (\hat{\mathbf{r}} \cdot \hat{\mathbf{r}}')^2]^{\frac{1}{2}}$.

FIG. 7: (a): Cross sections through the Coulomb hole $\rho_c(\mathbf{r}\mathbf{r}')$ in different directions corresponding to $\Theta = 0^\circ, 45^\circ, 90^\circ$ with respect to the center of the quantum dot - electron direction. The electron is at $r = 0.5$ a.u. as indicated by the arrow. (b): Surface plot of the Coulomb hole for an electron at the same position.

FIG. 8: The same as in Fig 7 except that the electron is at $r = 1$ a.u.

FIG. 9: The same as in Fig 7 except that the electron is at $r = 2$ a.u.

FIG. 10: The same as in Fig 7 except that the electron is at $r = 20$ a.u.

FIG. 11: The Hartree $\mathcal{E}_H(r)$, Pauli $\mathcal{E}_x(r)$, and Coulomb $\mathcal{E}_c(r)$ fields. The functions $2/r^2$ and $-1/r^2$ are also plotted in the asymptotic region. The Coulomb field decays asymptotically as $0(-10.1/r^4)$.

FIG. 12: (a): Kinetic ‘forces’ $z(r)$, $z_s(r)$ for the interacting and noninteracting systems, respectively. (b): The correlation-kinetic field $z_{tc}(r)$. Its asymptotic decay is of $0(9/r^3)$.

FIG. 13: A comparison of the Hartree $\mathcal{E}_H(r)$, Pauli $\mathcal{E}_x(r)$, Coulomb $\mathcal{E}_c(r)$, and correlation-kinetic $\mathcal{Z}_{tc}(r)$ fields.

FIG. 14: A comparison of the Hartree $W_H(r)$, Pauli $W_x(r)$, Coulomb $W_c(r)$, and correlation-kinetic $W_{t_c}(r)$ potentials.

FIG. 15: The potential $v_{ee}(r)$, and its electron-interaction $W_{ee}(r)$ and correlation-kinetic $W_{tc}(r)$ components.

APPENDIX A: QDFT EXPRESSIONS FOR THE 2^1S STATE OF A TWO-ELECTRON QUANTUM DOT IN A MAGNETIC FIELD

1. Electron density $\rho_1(\mathbf{r})$

$$\rho_1(r) = \frac{C_{01}^2}{2\Omega^4} \left\{ 4\pi e^{-\Omega r^2} (K_1 + L_1 r^2 + M_1 r^4 + N_1 r^6) + \right. \\ \left. \pi \sqrt{\pi \Omega} e^{-\frac{3}{2}\Omega r^2} \left[K_2 + L_2 r^2 + M_2 r^4 + N_2 r^6 \right] I_0(\Omega r^2/2) + \right. \\ \left. (L_3 r^2 + M_3 r^4 + N_3 r^6) I_1(\Omega r^2/2) \right\}, \quad (\text{A1})$$

where

$$\begin{aligned} C_{01} &= 0.1085631994, \\ \Omega &= \frac{2}{2.91199} = 0.686816, \\ K_1 &= 6B^2 + (2A^2 + 4B) \Omega + (1 + 2A) \Omega^2 + \Omega^3, \\ L_1 &= 18 B^2 \Omega + 4(A^2 + 2B) \Omega^2 + (1 + 2A) \Omega^3, \\ M_1 &= 9 B^2 \Omega^2 + (A^2 + 2B) \Omega^3, \\ N_1 &= B^2 \Omega^3, \\ K_2 &= 15AB + 6(A + B) \Omega + 4 \Omega^2, \\ L_2 &= 45AB \Omega + 12(A + B) \Omega^2 + 4 \Omega^3, \\ M_2 &= 28AB \Omega^2 + 4(A + B) \Omega^3, \\ N_2 &= 4AB \Omega^3, \\ L_3 &= 23AB \Omega + 8(A + B) \Omega^2 + 4 \Omega^3, \\ M_3 &= 24AB \Omega^2 + 4(A + B) \Omega^3, \\ N_3 &= 4AB \Omega^3, \\ A &= \frac{1}{4}(1 + \Omega - 2 \epsilon_r) = -0.2651111137, \\ B &= \frac{1}{9} \left(\frac{1}{4} - \frac{5\epsilon_r}{2} + \frac{9}{4}\Omega \right) = -0.1820822248, \\ \epsilon_r &= 1.37363 \end{aligned} \quad (\text{A2})$$

and $I_0(x)$ and $I_1(x)$ are the zeroth- and first-order modified Bessel functions [45].

The asymptotic structure of $\rho_1(\mathbf{r})$ near the center of the quantum dot, and in the classically

forbidden region, respectively, are as follows:

$$\rho_1(r) \underset{r \rightarrow 0}{\sim} 0.207 - 0.141 r^2 + 0.0753 r^4 + \dots \quad (\text{A3})$$

with $\rho_1(0) = 0.207299$ *a.u.*,

$$\rho_1(r) \underset{r \rightarrow \infty}{\sim} e^{-\Omega r^2} (-0.00171 r^3 + 0.00252 r^2 + 0.0549 r + 0.113 + \frac{0.0398}{r} + \frac{0.00902}{r^3} + \dots) \quad (\text{A4})$$

2. Pair-correlation density $g(\mathbf{r}\mathbf{r}')$

$$g(\mathbf{r}\mathbf{r}') = \frac{2 C_{01}^2 e^{-\Omega(r^2+r'^2)} \left[1 + |\mathbf{r} - \mathbf{r}'| + A|\mathbf{r} - \mathbf{r}'|^2 + B|\mathbf{r} - \mathbf{r}'|^3 \right]^2}{\rho_1(\mathbf{r})}, \quad (\text{A5})$$

where the constants C_{01}, Ω, A and B are given in Eq. (A2).

3. Single-particle density matrix $\gamma(\mathbf{r}\mathbf{r}')$

$$\gamma(\mathbf{r}\mathbf{r}') = 2 C_{01}^2 e^{-\Omega(r^2+r'^2)/2} \int e^{-\Omega y^2} \left[1 + |\mathbf{y} - \mathbf{r}| + A|\mathbf{y} - \mathbf{r}|^2 + B|\mathbf{y} - \mathbf{r}|^3 \right] \left[1 + |\mathbf{y} - \mathbf{r}'| + A|\mathbf{y} - \mathbf{r}'|^2 + B|\mathbf{y} - \mathbf{r}'|^3 \right] d\mathbf{y}, \quad (\text{A6})$$

where the constants C_{01}, Ω, A and B are given in Eq. (A2).

4. Dirac density matrix $\gamma_s(\mathbf{r}\mathbf{r}')$

$$\gamma_s(\mathbf{r}\mathbf{r}') = \sqrt{\rho_1(\mathbf{r}) \rho_1(\mathbf{r}')} \quad (\text{A7})$$

5. Electron-interaction field $\mathcal{E}_{ee}(\mathbf{r})$ and its components

$$\begin{aligned} \mathcal{E}_{ee}(\mathbf{r}) = & \frac{\pi C_{01}^2 e^{-2\Omega r^2}}{4 r \Omega^{5/2} \rho_1(\mathbf{r})} \left\{ \sqrt{\pi} r^2 e^{\Omega r^2/2} \left[I_1(\Omega r^2/2) \left(r^2(4A^2\Omega^2 + 14B^2\Omega + 8B\Omega^2) + 2A^2\Omega + 8A\Omega^2 + 4B^2r^4\Omega^2 + 4B\Omega - 8\Omega^3 + 4\Omega^2 + 3B^2 \right) \right. \right. \\ & \left. \left. + I_0(\Omega r^2/2) \left(r^2(4A^2\Omega^2 + 18B^2\Omega + 8B\Omega^2) + 6A^2\Omega + 8A\Omega^2 + 4B^2r^4\Omega^2 + 12B\Omega + 8\Omega^3 + 4\Omega^2 + 15B^2 \right) \right] \right. \\ & \left. + 16\sqrt{\Omega} \left[e^{\Omega r^2} \left(AB r^4 \Omega + r^2(A\Omega + B\Omega + 2AB) + \Omega \right) - \Omega \right] \right\}, \quad (\text{A8}) \end{aligned}$$

where the constants C_{01}, Ω, A and B are given in Eq. (A2).

The asymptotic structure of $\mathcal{E}_{ee}(\mathbf{r})$ and its Hartree $\mathcal{E}_H(\mathbf{r})$, Pauli-Coulomb $\mathcal{E}_{xc}(\mathbf{r})$, Pauli

$\mathcal{E}_x(\mathbf{r})$, and Coulomb $\mathcal{E}_c(\mathbf{r})$ components are

$$\begin{aligned}\mathcal{E}_{ee}(r) &\underset{r \rightarrow 0}{\sim} 0.955 r - 0.494 r^3, & \mathcal{E}_{ee}(r) &\underset{r \rightarrow \infty}{\sim} \frac{1}{r^2} - \frac{7.64}{r^4} + \frac{4.24}{r^5}, \\ \mathcal{E}_H(r) &\underset{r \rightarrow \infty}{\sim} \frac{2}{r^2} + \frac{4.92}{r^4}, & \mathcal{E}_{xc}(r) &\underset{r \rightarrow \infty}{\sim} -\frac{1}{r^2} - \frac{12.6}{r^4}, \\ \mathcal{E}_x(r) &\underset{r \rightarrow \infty}{\sim} -\frac{1}{r^2} - \frac{2.45}{r^4}, & \mathcal{E}_c(r) &\underset{r \rightarrow \infty}{\sim} -\frac{10.1}{r^4}.\end{aligned}\tag{A9}$$

6. Electron-interaction potential $W_{ee}(\mathbf{r})$ and its components

$$W_{ee}(\mathbf{r}) = - \int_{\infty}^r \mathcal{E}_{ee}(y) dy,\tag{A10}$$

where $\mathcal{E}_{ee}(r)$ is given in Eq. (A8). The Hartree potential is

$$\begin{aligned}W_H(\mathbf{r}) &= \int \frac{\rho_1(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' \\ &= 4 \int_0^r dr' \frac{r'}{r} \rho_1(r') K\left(\frac{r'^2}{r^2}\right) + 4 \int_r^\infty dr' \rho(r') K\left(\frac{r^2}{r'^2}\right),\end{aligned}\tag{A11}$$

where $K(x)$ is the complete elliptic integral of the first kind [45]. The asymptotic structure of $W_{ee}(r)$ and its Hartree $W_H(r)$, Pauli-Coulomb $W_{xc}(r)$, Pauli $W_x(r)$, and Coulomb $W_c(r)$ components are

$$\begin{aligned}W_{ee}(r) &\underset{r \rightarrow \infty}{\sim} \frac{1}{r} - \frac{2.55}{r^3}, & W_H(r) &\underset{r \rightarrow \infty}{\sim} \frac{2}{r} + \frac{1.64}{r^3}, \\ W_{xc}(r) &\underset{r \rightarrow \infty}{\sim} -\frac{1}{r} - \frac{4.18}{r^3}, & W_x(r) &\underset{r \rightarrow \infty}{\sim} -\frac{1}{r} - \frac{0.818}{r^3}, \\ W_c(r) &\underset{r \rightarrow \infty}{\sim} -\frac{3.37}{r^3},\end{aligned}\tag{A12}$$

7. Electron-interaction energy E_{ee}

$$\begin{aligned}E_{ee} &= \pi^2 C_{01}^2 \left[\sqrt{\frac{\pi}{2\Omega^9}} \left(\Omega (2A\Omega + \Omega^2 + \Omega + 3A^2 + 6B) + 15B^2 \right) + \right. \\ &\quad \left. 2 \left(2\Omega(A + B) + \Omega^2 + 8AB \right) / \Omega^4 \right] \\ &= 0.600476 \text{ a.u.},\end{aligned}\tag{A13}$$

where the constants C_{01}, Ω, A and B are given in Eq. (A2).

8. Kinetic energy tensor $t_{\alpha\beta}[\mathbf{r}; \gamma]$

$$t_{\alpha\beta}[\mathbf{r}; \gamma] = \frac{r_\alpha r_\beta}{r^2} f(r) + \delta_{\alpha\beta} k(r),\tag{A14}$$

where

$$f(r) = \pi C_{01}^2 e^{-2\Omega r^2} \left[\frac{r}{\Omega} \frac{\partial f_1(r)}{\partial r} - 2 r \frac{\partial f_2(r)}{\partial r} \right] + \frac{\Omega^2}{2} r^2 \rho_1(r), \quad (\text{A15})$$

$$k(r) = \frac{\pi C_{01}^2}{\Omega} e^{-2\Omega r^2} f_1(r), \quad (\text{A16})$$

$$f_1(r) = \frac{1}{2 \Omega^2 r^2} \left\{ -\Omega + e^{\Omega r^2} \left[\Omega + (4\Omega A^2 + 6\Omega B + 18A^2 + 9\Omega B^2 r^2) r^2 \right] + \right. \\ \left. A\sqrt{\pi\Omega} r^2 e^{\Omega r^2/2} \left[(2\Omega + 9B + 6\Omega B r^2) I_0(\Omega r^2/2) + (2\Omega + 3B + 6\Omega B r^2) I_1(\Omega r^2/2) \right] \right\}, \quad (\text{A17})$$

$$f_2(r) = \frac{e^{\Omega r^2/2}}{8\Omega^3} \left\{ 4e^{\Omega r^2/2} \left[(6B^2 + 2A^2\Omega + 4B\Omega + \Omega^2 + 2A\Omega^2) + \right. \right. \\ \left. (12B^2\Omega + 2A^2\Omega^2 + 4B\Omega^2) r^2 + 3B^2\Omega^2 r^4 \right] + \sqrt{\pi\Omega} \\ \left[\left(15AB + 6A\Omega + 6B\Omega + 4\Omega^2 + (30AB\Omega + 6(A+B) \right. \right. \\ \left. \left. \Omega^2) r^2 + 10AB\Omega^2 r^4 \right) I_0(\Omega r^2/2) + \left(2\Omega(10AB + \right. \right. \\ \left. \left. 3A\Omega + 3B\Omega) r^2 + 10AB\Omega^2 r^4 \right) I_1(\Omega r^2/2) \right] \right\}, \quad (\text{A18})$$

where the constants C_{01}, Ω, A and B are given in Eq. (A2).

9. Kinetic energy tensor $t_{s,\alpha\beta}[\mathbf{r}; \gamma_{\mathbf{s}}]$

$$t_{s,\alpha\beta}[\mathbf{r}; \gamma_{\mathbf{s}}] = \frac{r_\alpha r_\beta}{8r^2 \rho_1(r)} \left(\frac{\partial \rho_1(r)}{\partial r} \right)^2. \quad (\text{A19})$$

10. Kinetic ‘force’ $z_\alpha[\mathbf{r}; \gamma]$

$$z_\alpha[\mathbf{r}; \gamma] = \frac{2 r_\alpha}{r} \left\{ \frac{\partial [f(r) + k(r)]}{\partial r} + \frac{f(r)}{r} \right\}, \quad (\text{A20})$$

where the functions $f(r)$ and $k(r)$ are given in Eq. (A15) and (A16), respectively. The asymptotic structures are

$$z(r) \underset{r \rightarrow 0}{\sim} 0.703 r - r^3 + 0.459 r^5 + \dots \quad (\text{A21})$$

$$z(r) \underset{r \rightarrow \infty}{\sim} e^{-\Omega r^2} (-0.0417r^5 - 0.346r^4 - 0.215r^3 + 0.332r^2 + 0.202r \\ - 0.0772 + \frac{0.216}{r} + \frac{0.0343}{r^2} - \frac{0.0650}{r^4} + \dots), \quad (\text{A22})$$

where Ω is given in Eq. (A2).

11. Kinetic ‘force’ $z_{s,\alpha}[\mathbf{r}; \gamma_s]$

$$\begin{aligned} z_{s,\alpha}[\mathbf{r}; \gamma_s] &= 2 \sum_{\beta=1}^2 \nabla_{\beta} t_{s,\alpha\beta}[\mathbf{r}; \gamma_s] \\ &= \frac{r_{\alpha}}{2r\rho_1(r)} \frac{\partial \rho_1(r)}{\partial r} \left[-\frac{1}{2\rho_1(r)} \left(\frac{\partial \rho_1(r)}{\partial r} \right)^2 + \frac{\partial^2 \rho_1(r)}{\partial r^2} + \frac{1}{2r} \frac{\partial \rho_1(r)}{\partial r} \right], \end{aligned} \quad (\text{A23})$$

$$z_{s,\alpha}(r) \underset{r \rightarrow 0}{\sim} 0.287 r - 0.699 r^3 + 0.519 r^5 + \dots \quad (\text{A24})$$

$$\begin{aligned} z_{s,\alpha}(r) \underset{r \rightarrow \infty}{\sim} e^{-\Omega r^2} &(-0.757 r^5 + 2.46 r^4 - 3.50 r^3 + 3.90 r^2 + \\ &32.2 r - 76.8 + \frac{58.5}{r} + \frac{204}{r^2} - \frac{815}{r^3} + \dots, \end{aligned} \quad (\text{A25})$$

where Ω is given in Eq. (A2).

12. Correlation-kinetic field $\mathcal{Z}_{t_c}(\mathbf{r})$ and potential $W_{t_c}(\mathbf{r})$

$$\mathcal{Z}_{t_c}(\mathbf{r}) = \frac{z_s(r) - z(r)}{\rho_1(r)}, \quad \mathcal{Z}_{t_c}(r) \underset{r \rightarrow \infty}{\sim} \frac{9}{r^3} - \frac{20}{r^4} + \dots, \quad (\text{A26})$$

$$W_{t_c}(\mathbf{r}) = - \int_{\infty}^r \mathcal{Z}_{t_c}(y) dy, \quad W_{t_c}(r) \underset{r \rightarrow \infty}{\sim} \frac{9}{2 r^2} + \dots \quad (\text{A27})$$

13. Effective electron-interaction potential $v_{ee}(\mathbf{r})$

$$v_{ee}(r) = W_{ee}(r) + W_{t_c}(r), \quad (\text{A28})$$

$$v_{ee}(r) \underset{r \rightarrow 0}{\sim} 2.07 + 0.262 r^2 + \dots, \quad v_{ee}(r) \underset{r \rightarrow \infty}{\sim} \frac{1}{r} + \frac{9}{2 r^2} + \dots \quad (\text{A29})$$

14. Kinetic Energy T

$$\begin{aligned} T &= \int \int \Psi_{01}(\mathbf{r}\mathbf{R}) \left(\nabla_r^2 + \frac{1}{4} \nabla_R^2 \right) \Psi_{01}(\mathbf{r}\mathbf{R}) d\mathbf{r} d\mathbf{R} \\ &= \frac{\pi^2 C_{01}^2}{4\Omega^4} \left(16\Omega (2A^2 + 3B) + \sqrt{2\pi} \Omega^{3/2} (11A + 3B) + 57\sqrt{2\pi}\Omega AB \right. \\ &\quad \left. + 8(A+1)\Omega^2 + 3\sqrt{2\pi}\Omega^{5/2} + 4\Omega^3 + 192B^2 \right) \\ &= 1.266683 \text{ a.u.}, \end{aligned} \quad (\text{A30})$$

where the constants C_{01} , Ω , A and B are given in Eq. (A2).

15. External Energy E_{ext}

$$\begin{aligned}
E_{ext} &= \int \rho_1(\mathbf{r}) \left(\frac{1}{2} k_{\text{eff}} r^2 \right) d\mathbf{r} \\
&= \frac{\pi^2 C_{01}^2}{4\Omega^4} \left(64\Omega (A^2 + 2B) + 21\sqrt{2\pi} \Omega^{3/2}(A + B) + 135\sqrt{2\pi\Omega}AB + \right. \\
&\quad \left. 12(2A + 1)\Omega^2 + 5\sqrt{2\pi}\Omega^{5/2} + 4\Omega^3 + 480B^2 \right) \\
&= 1.566917 \text{ a.u.}, \tag{A31}
\end{aligned}$$

where the constants C_{01} , Ω , A and B are given in Eq. (A2), and $k_{\text{eff}} = \Omega^2 = 0.471716$.

16. Expectation values

$$\begin{aligned}
\langle r \rangle &= \int \rho_1(r) r d\mathbf{r} \\
&= \frac{\pi^{3/2} C_{01}^2}{8\Omega^{11/2}} \left[\pi \left(94\Omega (A^2 + 2B) + 20(2A + 1)\Omega^2 + 8\Omega^3 + 639B^2 \right) + \right. \\
&\quad \left. 4\sqrt{2} \Gamma\left(\frac{3}{4}\right)^2 \sqrt{\Omega} \left(15\Omega(A + B) + 4\Omega^2 + 87AB \right) + \right. \\
&\quad \left. \frac{32\sqrt{2\Omega}\pi^2 \left(7\Omega(A + B) + 2\Omega^2 + 40AB \right)}{\Gamma\left(-\frac{1}{4}\right)^2} \right] \\
&= 3.276784 \text{ a.u.}, \tag{A32}
\end{aligned}$$

where the constants C_{01} , Ω , A and B are given in Eq. (A2), and $\Gamma(x)$ is the Gamma function [45].

$$\begin{aligned}
\langle r^2 \rangle &= \int \rho_1(r) r^2 d\mathbf{r} \\
&= \frac{\pi^2 C_{01}^2}{2\Omega^6} \left(64\Omega (A^2 + 2B) + 21\sqrt{2\pi} \Omega^{3/2}(A + B) + 135\sqrt{2\pi\Omega}AB + \right. \\
&\quad \left. 12(2A + 1)\Omega^2 + 5\sqrt{2\pi}\Omega^{5/2} + 4\Omega^3 + 480B^2 \right) \\
&= 6.643480 \text{ a.u.}, \tag{A33}
\end{aligned}$$

where the constants C_{01}, Ω, A and B are given in Eq. (A2).

$$\begin{aligned}
\left\langle \frac{1}{r} \right\rangle &= \int \rho_1(r) \left(\frac{1}{r} \right) d\mathbf{r} \\
&= \frac{\pi^{3/2} C_{01}^2}{4\Omega^{9/2}} \left[\pi \left(\Omega (38A^2 + 76B) + 4\Omega^2(2\Omega + 6A + 3) + 189B^2 \right) + \right. \\
&\quad 4\sqrt{2} \Gamma\left(\frac{3}{4}\right)^2 \sqrt{\Omega} \left(6\Omega(A + B) + 2\Omega^2 + 27AB \right) + \\
&\quad \left. \frac{32\sqrt{2}\Omega\pi^2 \left(4\Omega(A + B) + 2\Omega^2 + 15AB \right)}{\Gamma\left(-\frac{1}{4}\right)^2} \right] \\
&= 1.930015 \text{ a.u.}, \tag{A34}
\end{aligned}$$

where the constants C_{01}, Ω, A and B are given in Eq. (A2), and $\Gamma(x)$ is the Gamma function [45].

$$\begin{aligned}
\langle \delta(\mathbf{r}) \rangle &= \rho_1(0) \\
&= \frac{C_{01}^2}{2\Omega^4} \left[4\pi \left((2A^2 + 4B)\Omega + (2A + 1)\Omega^2 + \Omega^3 + 6B^2 + \right. \right. \\
&\quad \left. \left. \pi^{3/2} \sqrt{\Omega} \left(6\Omega(A + B) + 4\Omega^2 + 15AB \right) \right) \right] \\
&= 0.207299 \text{ a.u.}, \tag{A35}
\end{aligned}$$

where the constants C_{01}, Ω, A and B are given in Eq. (A2).

APPENDIX B: DERIVATION OF THE KINETIC-ENERGY TENSOR AND KINETIC ‘FORCE’

The kinetic energy tensor $t_{\alpha\beta}[\mathbf{r}; \gamma]$ is defined as

$$t_{\alpha\beta}[\mathbf{r}; \gamma] = \frac{1}{4} \left[\frac{\partial^2}{\partial r_{p\alpha} \partial r_{q\beta}} + \frac{\partial^2}{\partial r_{p\beta} \partial r_{q\alpha}} \right] \gamma(\mathbf{r}_p \mathbf{r}_q) \Big|_{\mathbf{r}_p = \mathbf{r}_q = \mathbf{r}}, \tag{B1}$$

where the single-particle density matrix is

$$\gamma(\mathbf{r}_p \mathbf{r}_q) = 2 \int \Psi^*(\mathbf{r}_p \mathbf{r}_2) \Psi(\mathbf{r}_q \mathbf{r}_2) d\mathbf{r}_2. \tag{B2}$$

The spatial part of the first singlet excited state wave function is

$$\Psi_{01}(\mathbf{r}_1 \mathbf{r}_2) = C_{01} e^{-\Omega(r_1^2 + r_2^2)/2} \left[1 + |\mathbf{r}_2 - \mathbf{r}_1| + A|\mathbf{r}_2 - \mathbf{r}_1|^2 + B|\mathbf{r}_2 - \mathbf{r}_1|^3 \right], \tag{B3}$$

where the constants C_{01}, Ω, A and B are given in Eq.(A2). Due to the symmetry of $\mathbf{r}_q$ and $\mathbf{r}_p$ in $\gamma(\mathbf{r}_p \mathbf{r}_q)$ and the fact that the wave function is real, the tensor in (B1) reduces to

$$t_{\alpha\beta}[\mathbf{r}; \gamma] = \int \frac{\partial \Psi_{01}(\mathbf{r}_p \mathbf{r}_2)}{\partial r_{p\alpha}} \frac{\partial \Psi_{01}(\mathbf{r}_q \mathbf{r}_2)}{\partial r_{q\beta}} \Big|_{\mathbf{r}_p=\mathbf{r}_q=\mathbf{r}}. \quad (\text{B4})$$

Substituting the wave function of (B3) into (B4) and employing the relations

$$\frac{\partial |\mathbf{r}_2 - \mathbf{r}_p|}{\partial r_{p\alpha}} = -\frac{(r_{2\alpha} - r_{p\alpha})}{|\mathbf{r}_2 - \mathbf{r}_p|} = -\frac{(r_{2\alpha} - r_\alpha)}{|\mathbf{r}_2 - \mathbf{r}|}, \quad (\text{B5})$$

and

$$\frac{\partial}{\partial r_{p\alpha}} e^{-\Omega(r_p^2+r_2^2)/2} = -\Omega r_{p\alpha} e^{-\Omega(r_p^2+r_2^2)/2} = -\Omega r_\alpha e^{-\Omega(r^2+r_2^2)/2}, \quad (\text{B6})$$

and transforming the coordinates to $\mathbf{r}_3 = \mathbf{r}_2 - \mathbf{r}$, the resulting $t_{\alpha\beta}[\mathbf{r}; \gamma]$ is the sum of 4 terms, which are

$$\text{Term 1} = C_{01}^2 e^{-2\Omega r^2} \int r_{3\alpha} r_{3\beta} g_1^2(r_3) e^{-\Omega(r_3^2+2\mathbf{r}\cdot\mathbf{r}_3)} d\mathbf{r}_3, \quad (\text{B7})$$

$$\text{Term 2} = C_{01}^2 e^{-2\Omega r^2} (\Omega r_\beta) \int r_{3\alpha} g_1(r_3) g_0(r_3) e^{-\Omega(r_3^2+2\mathbf{r}\cdot\mathbf{r}_3)} d\mathbf{r}_3, \quad (\text{B8})$$

$$\text{Term 3} = C_{01}^2 e^{-2\Omega r^2} (\Omega r_\alpha) \int r_{3\beta} g_1(r_3) g_0(r_3) e^{-\Omega(r_3^2+2\mathbf{r}\cdot\mathbf{r}_3)} d\mathbf{r}_3, \quad (\text{B9})$$

$$\text{Term 4} = C_{01}^2 e^{-2\Omega r^2} (r_\alpha r_\beta \Omega^2) \int g_0^2(r_3) e^{-\Omega(r_3^2+2\mathbf{r}\cdot\mathbf{r}_3)} d\mathbf{r}_3, \quad (\text{B10})$$

where

$$g_0(r_3) = 1 + r_3 + A r_3^2 + B r_3^3, \quad (\text{B11})$$

and

$$g_1(r_3) = \frac{1}{r_3} + 2A + 3B r_3. \quad (\text{B12})$$

The vector-components $r_{3\alpha}$ and $r_{3\beta}$ in the integrand of (B7) to (B9) will be eliminated through the equalities

$$r_{3\alpha} e^{-2\Omega \mathbf{r}\cdot\mathbf{r}_3} = -\frac{1}{2\Omega} \frac{\partial}{\partial r_\alpha} e^{-2\Omega \mathbf{r}\cdot\mathbf{r}_3}, \quad (\text{B13})$$

and

$$r_{3\beta} e^{-2\Omega \mathbf{r}\cdot\mathbf{r}_3} = -\frac{1}{2\Omega} \frac{\partial}{\partial r_\beta} e^{-2\Omega \mathbf{r}\cdot\mathbf{r}_3}, \quad (\text{B14})$$

and then by first evaluating the $d\theta_3$ integral, Term 1 in (B7) reduces to

$$\text{Term 1} = \frac{2\pi C_{01}^2}{4\Omega^2} e^{-2\Omega r^2} \frac{\partial^2}{\partial r_\alpha \partial r_\beta} \int_0^\infty r_3 g_1^2(r_3) e^{-\Omega r_3^2} I_0(2\Omega r r_3) dr_3, \quad (\text{B15})$$

where $I_0(x)$ is the zeroth-order modified Bessel function [45].

Since the lowest order of g_1 is $1/r_3$, the entire integrand of (B14) has the lowest order of $1/r_3$, which is singular at $r_3 = 0$. In order to eliminate the singularity, we employ

$$\frac{\partial}{\partial r_\beta} I_0(2\Omega r r_3) = \frac{2\Omega r_\beta r_3}{r} I_1(2\Omega r r_3), \quad (\text{B16})$$

where $I_1(x)$ is the first-order modified Bessel function [45].

Then by evaluating the dr_3 integral, and employing the equality for a general function $y(r)$ as follows

$$\frac{\partial}{\partial r_\alpha} [r_\beta y(r)] = \delta_{\alpha\beta} y(r) + \frac{r_\alpha r_\beta}{r} \frac{\partial y(r)}{\partial r}, \quad (\text{B17})$$

the Term 1 in (B15) reduces to

$$\text{Term 1} = \frac{\pi C_{01}^2}{\Omega} e^{-2\Omega r^2} \left[\delta_{\alpha\beta} f_1(r) + \frac{r_\alpha r_\beta}{r} \frac{\partial f_1(r)}{\partial r} \right], \quad (\text{B18})$$

where $f_1(r)$ is given in (A17).

In order to evaluate Term 2 and Term 3 in (B8) and (B9), which are identical, we apply the equalities (B13) and (B14), evaluate the $d\theta_3$ integral first (no singularity in this case), then evaluate the dr_3 integral, and employ the following equality for any function $y(r)$

$$r_\beta \frac{\partial y(r)}{\partial r_\alpha} = \frac{r_\alpha r_\beta}{r} \frac{\partial y(r)}{\partial r}. \quad (\text{B19})$$

Therefore, by adding up Term 2 + Term 3 we obtain

$$\text{Term 2} + \text{Term 3} = -2\pi C_{01}^2 e^{-2\Omega r^2} \left(\frac{r_\alpha r_\beta}{r} \right) \frac{\partial f_2(r)}{\partial r} \quad (\text{B20})$$

where $f_2(r)$ is given in (A18).

With regard to Term 4, except $(r_\alpha r_\beta \Omega^2)$, the rest of the expression is equal to $1/2$ of the electron density $\rho_1(r)$ given in (A1). [Transform the coordinates back from $\mathbf{r}_3$ to $(\mathbf{r}_2 - \mathbf{r})$], thus

$$\text{Term 4} = \frac{r_\alpha r_\beta \Omega^2}{2} \rho_1(r). \quad (\text{B21})$$

On summing all Term 1 + Term 2 + Term 3 + Term 4 in (B18), (B20), and (B21), we obtain the kinetic energy density tensor $t_{\alpha\beta}[\mathbf{r}; \gamma]$ as given in (A14).

The component of the kinetic ‘force’ is defined as

$$z_\alpha[\mathbf{r}; \gamma] = 2 \sum_{\beta=1}^2 \nabla_\beta t_{\alpha\beta}[\mathbf{r}; \gamma]. \quad (\text{B22})$$

Upon substituting $t_{\alpha\beta}[\mathbf{r}; \gamma]$ in (A14) into (B22) we obtain

$$z_{\alpha}[\mathbf{r}; \gamma] = 2 \sum_{\beta=1}^2 \frac{\partial}{\partial r_{\beta}} \left[\frac{r_{\alpha} r_{\beta}}{r^2} f(r) + \delta_{\alpha\beta} k(r) \right], \quad (\text{B23})$$

where $f(r)$ and $k(r)$ are given in (A15) and (A16).

For the 2-Dimensional coordinate system, it can be shown that

$$\sum_{\beta=1}^2 \frac{\partial}{\partial r_{\beta}} (r_{\alpha} r_{\beta}) = 3 r_{\alpha}, \quad (\text{B24})$$

and in general, the following equalities hold

$$\sum_{\beta=1}^2 r_{\alpha} r_{\beta} \frac{\partial}{\partial r_{\beta}} \left[\frac{f(r)}{r^2} \right] = r_{\alpha} \left[\frac{1}{r} \frac{\partial f(r)}{\partial r} - \frac{2 f(r)}{r^2} \right], \quad (\text{B25})$$

$$\text{and } \sum_{\beta=1}^2 \frac{\partial}{\partial r_{\beta}} \left[\delta_{\alpha\beta} k(r) \right] = \frac{r_{\alpha}}{r} \frac{\partial k(r)}{\partial r}. \quad (\text{B26})$$

Finally, by substituting (B24), (B25) and (B26) into (B23), we obtain the components of the kinetic ‘force’ $z_{\alpha}[\mathbf{r}; \gamma]$ as given in (A20).

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