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Schrödinger-Pauli Theory of Electrons: New Perspectives

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Abstract

The Schrödinger-Pauli (SP) theory of electrons in an electromagnetic field explicitly accounts for the electron spin moment. The many-electron theory is complemented via a new descriptive perspective *viz.* that of the *individual electron* via its equation of motion or ‘Quantal Newtonian’ first law. The law is in terms of ‘classical’ fields whose sources are quantum mechanical expectation values of Hermitian operators taken with respect to the system wave function. The law states that *each electron* experiences an external and an internal field, the sum of which vanish. The external field is the sum of the binding electrostatic and a Lorentz field. The internal field is a sum of fields representative respectively of electron correlations due to the Pauli principle and Coulomb repulsion; the electron density; kinetic effects; and an internal magnetic field. The energy can be expressed in integral virial form in terms of these fields. The law is elucidated by application to the 2^3S state of a quantum dot in a magnetic field. It is proved that the Hamiltonian is an exactly known and universal functional of the wave function. This generalizes the SP equation, and reveals that its eigenfunctions and eigenvalues may be determined self-consistently. A Quantal density functional theory (QDFT) of the SP system is developed whereby additional properties are determined. A physical interpretation of Spin-DFT based on the QDFT mapping is provided. Further generalizations of the present work to the temporal case, and relativistic Dirac theory, are proposed.

I. INTRODUCTION

The Schrödinger-Pauli (SP) theory [1] is a description of a quantum-mechanical system comprised of N electrons in the presence of an external electromagnetic field: $\mathcal{E}(\mathbf{r}) = -\nabla v(\mathbf{r})/e$; $\mathcal{B}(\mathbf{r}) = \nabla \times A(\mathbf{r})$, where $v(\mathbf{r})$ and $A(\mathbf{r})$ are scalar electrostatic and vector magnetic potentials. The theory is a generalization of Schrödinger theory [2] in that the electrons now additionally possess a spin moment. Thus, the interaction of the magnetic field with both the orbital and spin angular momentum is explicitly considered. Another factor of significance in the context of the present work is that in addition to the paramagnetic and diamagnetic components of the physical current density, there now exists a magnetization (spin) component. Originally, the corresponding non-relativistic SP Hamiltonian was *ad hoc*. Soon thereafter it was shown that the equation could be obtained as the non-relativistic limit of the Dirac equation [3]. Many years later Feynman noted that the Hamiltonian could be derived non-relativistically by what we refer to as the Feynman kinetic energy operator [4].

Our present understanding of SP theory is the traditional Copenhagen interpretation [5] of quantum mechanics. In addition to the Born rule that interprets the many-electron wave function $\Psi(\mathbf{X})$ as a ‘probability amplitude’, this includes the Heisenberg uncertainty principle; the information content that $\Psi(\mathbf{X})$ incorporates all the possible information about a quantum state; the wave function collapse, *i.e.* when a measurement is made, the state collapses to an eigenstate of the Hermitian operator associated with the observable being measured; the correspondence principle; and the wave-particle complementarity. The wave function solutions of the SP equation are descriptive of the *N-electron* system. It is in this manner that the SP theory has been interpreted and employed since its advent.

The purpose of this paper is to describe SP theory of the many-electron system from a *new* perspective. The perspective falls within the umbrella of the statistical Copenhagen interpretation, but is one that leads to further physical and mathematical insights into the system and eigenvalue equation. The perspective is that of the *individual electron* via its stationary-state equation of motion or ‘Quantal Newtonian’ first law (QNFL). *The law is derived from the SP equation, and is exact.* The law is a description in terms of ‘classical’ fields whose sources are quantum-mechanical expectations of Hermitian operators taken with respect to the wave function $\Psi(\mathbf{X})$. The QNFL thus adheres to the probabilistic interpretation of quantum mechanics.

The fields experienced by each electron obey a classical physics equation. Hence, as in classical physics, the fields experienced by each electron pervade all space, and are thus tangible in the classical sense. In that context, the fields are deterministic. The existence of fields, other than the external fields, as experienced by each electron, is a new understanding. The total energy E of the many-electron system is obtained from these fields. From a mathematical perspective, it is further proved that the Hamiltonian is an *exactly known* and *universal* functional of the wave function. This then generalizes the SP eigenvalue equation. The generalized form of the equation in turn exhibits its intrinsic self-consistent nature. What this means, as will be explained, is that the eigenfunctions and eigenvalues can be determined in a self-consistent manner. Next, the interacting system of electrons with spin as described by the SP equation is mapped via quantal density functional theory (QDFT) [6, 7] to one of *noninteracting* fermions possessing the same electronic density $\rho(\mathbf{r})$ and physical current density $\mathbf{j}(\mathbf{r})$, and from which the total energy E can be obtained. The mapping provides a rigorous *physical* interpretation of the model system. The mapping allows for the determination of additional properties of the system not obtainable solely by solution of the SP equation. The noninteracting fermion model also constitutes an alternative description of the physical system. It is a *local effective potential* theory, and as such it is more amenable to numerical solution. As the mapping to the model system explicitly accounts for the electron spin moment, it leads to physical insights into other local effective potential theories involving electron spin. In particular, we provide a physical understanding of the electron-interaction energy functional and functional derivative of spin density functional theory (SDFT). The SDFT we consider [8–10] is of the system of electrons possessing spin in the presence of a static electromagnetic field. The above perspectives thus constitute a complement to our present day understanding of SP theory.

(Prior to commencing, we clarify that the perspective being presented is *not* a *one-electron* theory as in Hartree-Fock [11, 12] or local effective potential theories [6–10]. It is a *many-electron* theory in which the QNFL is descriptive of the fields experienced by *each electron* in the sea of electrons.)

The non-relativistic SP Hamiltonian \hat{H} for spin $\frac{1}{2}$ particles is the sum of the Feynman [4] kinetic \hat{T}_F , electron-interaction potential \hat{W} , and external electrostatic potential \hat{V} op-

erators. In atomic units (charge of electron $-e$, $e = \hbar = m = 1$) the Hamiltonian is

$$\hat{H} = \hat{T}_F + \hat{W} + \hat{V}, \quad (1)$$

where

$$\hat{T}_F = \frac{1}{2} \sum_k (\boldsymbol{\sigma}_k \cdot \hat{\mathbf{p}}_{k,phys}) (\boldsymbol{\sigma}_k \cdot \hat{\mathbf{p}}_{k,phys}), \quad (2)$$

$$\hat{W} = \frac{1}{2} \sum'_{k,\ell} \frac{1}{|\mathbf{r}_k - \mathbf{r}_\ell|}, \quad (3)$$

$$\hat{V} = \sum_k v(\mathbf{r}_k). \quad (4)$$

Here the physical momentum operator $\hat{\mathbf{p}}_{phys} = (\hat{\mathbf{p}} + \frac{1}{c} \mathbf{A}(\mathbf{r}))$, with $\hat{\mathbf{p}} = -i\nabla$ the canonical momentum operator. The $\boldsymbol{\sigma}$ is the Pauli spin matrix: $\mathbf{s} = \frac{1}{2} \boldsymbol{\sigma}$, with \mathbf{s} the electron spin angular momentum vector operator. The external scalar electrostatic potential $v(\mathbf{r})$ corresponds to the binding potential of the electrons. For natural atoms, molecules and solids, it is the Coulomb potential due to the positively charged nucleus. For metals and metal surfaces in the jellium model approximation, it corresponds to the potential due to the positively charged jellium background. For semiconductor quantum dots, the binding potential is harmonic. On substituting for $\hat{\mathbf{p}}_{phys}$ and $\boldsymbol{\sigma}$ in the kinetic energy operator equation, the Hamiltonian \hat{H} may be written as

$$\hat{H} = \frac{1}{2} \sum_k \left(\hat{\mathbf{p}}_k + \frac{1}{c} \mathbf{A}(\mathbf{r}_k) \right)^2 + \frac{1}{c} \sum_k \mathcal{B}(\mathbf{r}_k) \cdot \mathbf{s}_k + \hat{W} + \hat{V}, \quad (5)$$

which then indicates the interaction of the magnetic field with both the orbital and spin moment of the electrons. It is interesting to note that both interactions arise via the Feynman kinetic energy operator T_F . The spin magnetic moment obtained this way has the correct gyromagnetic ratio $g = 2$. The corresponding SP equation is then

$$\hat{H}\Psi(\mathbf{X}) = E\Psi(\mathbf{X}), \quad (6)$$

with $\{\Psi(\mathbf{X}), E\}$ the eigenfunctions and eigenvalues; $\mathbf{X} = \mathbf{x}, \dots, \mathbf{x}_N$; $\mathbf{x} = \mathbf{r}\sigma$; and $\mathbf{r}\sigma$ the spatial and spin coordinates.

There are five components to the paper:

1. The first is comprised of the description of the quantum-mechanical system as defined by the Hamiltonian \hat{H} of Eq. (5) in terms of the ‘classical’ fields that satisfy the corresponding QNFL. This description is valid for *arbitrary state* whether ground, excited

or degenerate. (The derivation of the law which now explicitly accounts for electron spin is given in Appendix A.)

The statement of the QNFL is that the sum of the external $\mathcal{F}^{\text{ext}}(\mathbf{r})$ and internal $\mathcal{F}^{\text{int}}(\mathbf{r})$ fields experienced by *each* electron vanishes. The external field $\mathcal{F}^{\text{ext}}(\mathbf{r})$ is a sum of the external binding electrostatic $\mathcal{E}(\mathbf{r})$ and the Lorentz $\mathcal{L}(\mathbf{r})$ fields. The Lorentz field $\mathcal{L}(\mathbf{r})$ depends upon the cross-product of the physical current density $\mathbf{j}(\mathbf{r})$ and the magnetic field $\mathcal{B}(\mathbf{r})$. Thus, in the QNFL, the Lorentz field $\mathcal{L}(\mathbf{r})$, derived from a quantum-mechanical Lorentz ‘force’, appears *explicitly*.

The internal field $\mathcal{F}^{\text{int}}(\mathbf{r})$ is a sum of fields, each representative of a property of the system: properties such as the correlations due to the Pauli principle and Coulomb repulsion, the field being $\mathcal{E}_{ee}(\mathbf{r})$; the kinetic effects, $\mathcal{Z}(\mathbf{r})$; the electron density, $\mathcal{D}(\mathbf{r})$; and an internal magnetic field $\mathcal{I}_m(\mathbf{r})$ component.

One significant feature of the law is that in addition to the expected external electrostatic and Lorentz fields, each electron also experiences an internal field. And that these fields are representative of the intrinsic properties of the system. Whilst one might expect an internal field representative of Coulombic and Pauli principle electron-electron repulsion, one learns that there exist other components of the internal field. Hence, there exists a field representative of kinetic effects, and one representative of the electron density. And, that there also exists an internal magnetic field component.

The magnetic field contributions to the QNFL are the external Lorentz $\mathcal{L}(\mathbf{r})$ and internal magnetic $\mathcal{I}_m(\mathbf{r})$ fields. Provided the sum of these fields is conservative, it is then possible to define a scalar (path-independent) magnetic potential $v_m(\mathbf{r})$ in a manner similar to the external scalar electrostatic potential $v(\mathbf{r})$.

The QNFL is a *sum rule*. As such it can be employed as a constraint applied to approximate wave functions or as a test of the accuracy of such wave functions.

2. To elucidate the ideas underlying the quantal-source field perspective, we apply [13] them to the triplet 2^3S state of a 2-electron 2-dimensional quantum dot or ‘artificial atom’ in a magnetic field. Quantum dots [14, 15] are a fertile area for study of the electronic structure of reduced dimensional systems because their size and state can be manipulated by electric and magnetic fields. In this example, the Pauli term is finite, and the interaction of the magnetic field with the spin moment explicitly accounted for. In

particular we demonstrate the satisfaction of the QNFL by the various fields experienced by each electron. Additionally, we obtain the total energy E of the quantum dot via the fields.

3. The third component is a generalization of the SP equation which shows the Hamiltonian \hat{H} to be a functional of the wave function Ψ , i.e. $\hat{H} = \hat{H}[\Psi]$. Hence, the SP equation can be written in a more general form as

$$\hat{H}[\Psi]\Psi(\mathbf{X}) = E[\Psi]\Psi(\mathbf{X}). \quad (7)$$

In Eq. (7) the fact that the eigenvalues E too are functionals of the wave function Ψ is also explicitly indicated. The generalization of the SP equation is a consequence of the QNFL. It is therefore valid for *arbitrary* state. As the first law is in terms of fields whose sources are quantum-mechanical expectations of Hermitian operators taken with respect to the wave function Ψ , the functional $\hat{H}[\Psi]$ is *exactly known*. The functional $\hat{H}[\Psi]$ is also *universal* in that it is valid for any electronic system. It is evident from the generalized form of the SP equation that it is intrinsically a self-consistent eigenvalue equation. As will be explained, given the SP Hamiltonian of a system, its eigenfunctions and eigenvalues can be determined self-consistently. In the self-consistent procedure, the eigenvalue E at each iteration depends upon the solution of the equation for that iteration. It is for this reason that the eigenvalue E is written as the functional $E[\Psi]$. (The generalized SP equation is akin to the Hartree-Fock theory [11, 12] equations in which the corresponding Hamiltonian \hat{H}^{HF} is a functional of the single-particle spin-orbitals $\phi_i(\mathbf{x})$, i.e. $\hat{H}^{\text{HF}} = \hat{H}^{\text{HF}}[\phi_i(\mathbf{x})]$. The Hartree-Fock theory equations are then $\hat{H}^{\text{HF}}[\phi_i]\phi_i(\mathbf{x}) = \epsilon_i\phi_i(\mathbf{x})$, where the ϵ_i are the eigenvalues. These equations are then solved self-consistently.)

4. The fourth component of the paper constitutes the mapping of the interacting system of electrons as defined by the SP equation of Eq. (6) to one of noninteracting fermions possessing the same *basic variables* of the density $\rho(\mathbf{r})$ and physical current density $\mathbf{j}(\mathbf{r})$. The further constraints of the mapping are that of fixed electron number N , and total orbital \mathbf{L} and spin \mathbf{S} angular momentum. (Basic variables in quantum mechanics are gauge invariant properties, knowledge of which uniquely determine the external scalar and vector potentials to within a constant and gradient of a scalar function, respectively.) The mapping is accomplished via quantal density functional theory (QDFT). The mapping is

valid for *arbitrary* state of the interacting system. The state of the model system is also *arbitrary* provided the constraints are satisfied. The reasons for this mapping are twofold:

(a) The mapping to the model system allows for the determination of properties of the quantum system not obtainable solely via the solution of the SP equation. Such a property is the contribution of electron correlations due to the Pauli principle and Coulomb repulsion to the kinetic energy – the correlation-kinetic energy. Further, as a consequence of the mapping, it is also possible to separate the contributions to the total energy of the correlations due to the Pauli principle and Coulomb repulsion. Note that these properties are for the same $\{\rho, \mathbf{j}\}$ as those of the interacting system. The highest occupied eigenvalue of the corresponding differential equation is the negative of the ionization potential. The model system is thus also a complement to SP theory.

(b) The model system constitutes an alternate description of the physical system. Further, the equations governing the noninteracting fermions are easier to solve numerically. The corresponding ‘wave function’ is a Slater determinant of the model fermion spin-orbitals. The QDFT mapping provides the precise *physical* definition of the local effective potential in which all the many-body effects are incorporated. It is the work done by the model fermion in a *conservative* effective field. This local potential then generates the interacting system densities $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$.

The rationale for the choice of the densities $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ as the basic variables in the mapping stems from the ground state theorem of Hohenberg-Kohn (HK) [16] and of its extension by Pan-Sahni (PS) [17] to the presence of a uniform magnetic field. For a system of N electrons in an external electrostatic field $\mathcal{E}(\mathbf{r}) = -\nabla v(\mathbf{r})/e$, and in a nondegenerate ground state, HK proved that knowledge of the ground state density $\rho(\mathbf{r})$ uniquely determined the external scalar potential $v(\mathbf{r})$ to within a constant. The constraint in the proof is that of fixed electron number N . Hence, the nondegenerate ground state density $\rho(\mathbf{r})$ constitutes a basic variable. What PS proved was that in the added presence of a *uniform*

magnetostatic field $\mathcal{B}(\mathbf{r}) = \nabla \times \mathbf{A}(\mathbf{r})$, knowledge of the nondegenerate ground state $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ uniquely determines the potentials $\{v(\mathbf{r}), \mathbf{A}(\mathbf{r})\}$ to within a constant and gradient of a scalar function, respectively. The constraints in the proof are that of fixed electron number N , orbital \mathbf{L} , and spin \mathbf{S} angular momentum. The PS proof was for both spinless electrons and electrons with spin. Hence, in the presence of a magnetostatic field, the nondegenerate ground state $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ constitute basic variables. (The HK and PS proofs differ. There is a fundamental reason for this. In HK the relationship between $v(\mathbf{r})$ and the nondegenerate ground state Ψ is proved to be bijective or *one-to-one*. In the presence of a magnetic field, however, the relationship between $\{v(\mathbf{r}), \mathbf{A}(\mathbf{r})\}$ and the nondegenerate ground state Ψ is *many-to-one* and can be *infinite-to-one*. PS explicitly account for this many-to-one relationship, and in doing so, the proof follows a different path.)

5. In the final component of the paper, we employ the QDFT understanding of local effective potential theory of electrons with spin to provide a physical explanation of the energy functional of the spin-polarized densities and of the corresponding functional derivatives of SDFT. To facilitate this and to put the explanation in context, we provide a brief review of the foundations of SDFT.

The present work is a generalization to SP theory of ideas [6, 18–20] applicable to Schrödinger theory, thereby incorporating the added complexity of electron spin. *A priori* such an extension is not self-evident, and every facet must be rigorously proved. As such we derive the most general form of the QNFL for an electron with spin in the presence of a static electromagnetic field.

In Sect. II, we present the quantal-source-field perspective of the SP theory, and describe the new physical and mathematical insights as obtained from the QNFL. These ideas are then explicated in Sect. III by application to the triplet 2^3S state of a quantum dot in a magnetic field. The generalization of the SP equation whence it is proved that the Hamiltonian \hat{H} is a functional of the wave function Ψ , and that thereby the equation may be solved in a self-consistent manner, is discussed in Sect. IV. In Sect. V the local effective potential QDFT description of SP theory is developed. A physical interpretation of SDFT

based on this QDFT description is provided in Sect. VI. Finally, in Sect. VII, we summarize the conclusions of the work, and propose further generalizations to the time-dependent SP theory and to relativistic quantum mechanics via the Dirac theory.

II. DESCRIPTION IN TERMS OF QUANTAL SOURCES AND FIELDS: THE ‘QUANTAL NEWTONIAN’ FIRST LAW

In this section the quantum-mechanical system defined by the SP Hamiltonian is described in terms of ‘classical’ fields as experienced by each electron. The fields satisfy the QNFL – the equation of motion of the individual electron. The fields can be separated into two categories: an external $\mathcal{F}^{\text{ext}}(\mathbf{r})$ and an internal $\mathcal{F}^{\text{int}}(\mathbf{r})$ field. To define these fields, the many-electron Hamiltonian \hat{H} of Eq. (5) is rewritten in terms of the components of the physical current density operator $\hat{\mathbf{j}}(\mathbf{r})$ as

$$\begin{aligned} \hat{H} = \hat{T} + \frac{1}{c} \int \hat{\mathbf{j}}_p(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) d\mathbf{r} + \frac{1}{2c} \int \hat{\mathbf{j}}_d(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) d\mathbf{r} \\ + \frac{1}{c} \int \hat{\mathbf{j}}_m(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) d\mathbf{r} + \hat{W} + \hat{V}. \end{aligned} \quad (8)$$

In the above equation, \hat{T} is the canonical kinetic energy operator:

$$\hat{T} = \frac{1}{2} \sum_k \hat{p}_k^2, \quad (9)$$

and where the paramagnetic $\hat{\mathbf{j}}_p(\mathbf{r})$, diamagnetic $\hat{\mathbf{j}}_d(\mathbf{r})$, and magnetization $\hat{\mathbf{j}}_m(\mathbf{r})$ current density component operators are defined as

$$\hat{\mathbf{j}}_p(\mathbf{r}) = \frac{1}{2} \sum_k [\hat{\mathbf{p}}_k \delta(\mathbf{r}_k - \mathbf{r}) + \delta(\mathbf{r}_k - \mathbf{r}) \hat{\mathbf{p}}_k], \quad (10)$$

$$\hat{\mathbf{j}}_d(\mathbf{r}) = \frac{1}{c} \hat{\rho}(\mathbf{r}) \mathbf{A}(\mathbf{r}), \quad (11)$$

and

$$\hat{\mathbf{j}}_m(\mathbf{r}) = -c \nabla \times \hat{\mathbf{m}}(\mathbf{r}). \quad (12)$$

In turn the electronic density $\hat{\rho}(\mathbf{r})$ and magnetization density $\hat{\mathbf{m}}(\mathbf{r})$ operators of these equations are defined as

$$\hat{\rho}(\mathbf{r}) = \sum_k \delta(\mathbf{r}_k - \mathbf{r}), \quad (13)$$

and

$$\hat{\mathbf{m}}(\mathbf{r}) = -\frac{1}{c} \sum_k \mathbf{s}_k \delta(\mathbf{r}_k - \mathbf{r}). \quad (14)$$

The physical current density operator $\hat{\mathbf{j}}(\mathbf{r})$ is then obtained via its definition as

$$\hat{\mathbf{j}}(\mathbf{r}) = c \frac{\partial \hat{H}}{\partial \mathbf{A}(\mathbf{r})} = \hat{\mathbf{j}}_p(\mathbf{r}) + \hat{\mathbf{j}}_d(\mathbf{r}) + \hat{\mathbf{j}}_m(\mathbf{r}). \quad (15)$$

In terms of the density $\hat{\rho}(\mathbf{r})$ and current density $\hat{\mathbf{j}}(\mathbf{r})$ operators, the Hamiltonian \hat{H} of Eq. (8) may be written as

$$\hat{H} = \hat{T} + \frac{1}{c} \int \hat{\mathbf{j}}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) d\mathbf{r} - \frac{1}{2c^2} \int \hat{\rho}(\mathbf{r}) A^2(\mathbf{r}) d\mathbf{r} + \hat{W} + \hat{V}, \quad (16)$$

which then emphasizes the significance of both the electronic and physical current densities to the quantum system. Written in this manner facilitates the derivation of the QNFL (see Appendix A).

A. External Field $\mathcal{F}^{\text{ext}}(\mathbf{r})$

The external field $\mathcal{F}^{\text{ext}}(\mathbf{r})$ experienced by each electron is the sum of the binding electrostatic $\mathcal{E}(\mathbf{r})$ and Lorentz $\mathcal{L}(\mathbf{r})$ fields:

$$\mathcal{F}^{\text{ext}}(\mathbf{r}) = \mathcal{E}(\mathbf{r}) - \mathcal{L}(\mathbf{r}) = -\nabla v(\mathbf{r}) - \mathcal{L}(\mathbf{r}), \quad (17)$$

where the Lorentz field $\mathcal{L}(\mathbf{r})$ is defined in terms of the Lorentz ‘force’ $\boldsymbol{\ell}(\mathbf{r})$ and electronic density $\rho(\mathbf{r})$ (charge) as

$$\mathcal{L}(\mathbf{r}) = \frac{\boldsymbol{\ell}(\mathbf{r})}{\rho(\mathbf{r})}, \quad (18)$$

with

$$\boldsymbol{\ell}(\mathbf{r}) = \mathbf{j}(\mathbf{r}) \times \mathcal{B}(\mathbf{r}). \quad (19)$$

The electronic $\rho(\mathbf{r})$ and physical current $\mathbf{j}(\mathbf{r})$ densities are, respectively, the expectation values of the operators $\hat{\rho}(\mathbf{r})$ and $\hat{\mathbf{j}}(\mathbf{r})$, *i.e.* $\rho(\mathbf{r}) = \langle \Psi(\mathbf{X}) | \hat{\rho}(\mathbf{r}) | \Psi(\mathbf{X}) \rangle$, and $\mathbf{j}(\mathbf{r}) = \langle \Psi(\mathbf{X}) | \hat{\mathbf{j}}(\mathbf{r}) | \Psi(\mathbf{X}) \rangle$.

B. Internal Field $\mathcal{F}^{\text{int}}(\mathbf{r})$

The internal field $\mathcal{F}^{\text{int}}(\mathbf{r})$ is a sum of components each descriptive of a property of the system: an electron-interaction field $\mathcal{E}_{\text{ee}}(\mathbf{r})$; a kinetic field $\mathcal{Z}(\mathbf{r})$; the differential density

field $\mathcal{D}(\mathbf{r})$; and finally an internal magnetic field component $\mathcal{I}_m(\mathbf{r})$. Thus,

$$\mathcal{F}^{\text{int}}(\mathbf{r}) = \mathcal{E}_{\text{ee}}(\mathbf{r}) - \mathcal{Z}(\mathbf{r}) - \mathcal{D}(\mathbf{r}) - \mathcal{I}_m(\mathbf{r}). \quad (20)$$

The component fields and their respective quantal sources are defined next.

The electron-interaction field $\mathcal{E}_{\text{ee}}(\mathbf{r})$ in terms of the electron-interaction ‘force’ $\mathbf{e}_{\text{ee}}(\mathbf{r})$ and density $\rho(\mathbf{r})$ (charge) is

$$\mathcal{E}_{\text{ee}}(\mathbf{r}) = \frac{\mathbf{e}_{\text{ee}}(\mathbf{r})}{\rho(\mathbf{r})}, \quad (21)$$

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

$$\mathbf{e}_{\text{ee}}(\mathbf{r}) = \int \frac{P(\mathbf{r}\mathbf{r}')(\mathbf{r} - \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|^3} d\mathbf{r}'. \quad (22)$$

with $P(\mathbf{r}\mathbf{r}')$ the expectation value $P(\mathbf{r}\mathbf{r}') = \langle \Psi(\mathbf{X}) | \hat{P}(\mathbf{r}\mathbf{r}') | \Psi(\mathbf{X}) \rangle$, of the pair-correlation operator

$$\hat{P}(\mathbf{r}\mathbf{r}') = \sum_{k,\ell} \delta(\mathbf{r}_k - \mathbf{r}) \delta(\mathbf{r}_\ell - \mathbf{r}'). \quad (23)$$

The electron-interaction field $\mathcal{E}_{\text{ee}}(\mathbf{r})$ may equivalently be thought of as arising via Coulomb’s law from the quantal source of the pair-correlation density $g(\mathbf{r}\mathbf{r}') = P(\mathbf{r}\mathbf{r}')/\rho(\mathbf{r})$. The pair-correlation density can be separated into its *local* $\rho(\mathbf{r}')$ and *nonlocal* $\rho_{xc}(\mathbf{r}\mathbf{r}')$ components: $g(\mathbf{r}\mathbf{r}') = \rho(\mathbf{r}') + \rho_{xc}(\mathbf{r}\mathbf{r}')$, where $\rho_{xc}(\mathbf{r}\mathbf{r}')$ is the quantum-mechanical Fermi-Coulomb hole charge distribution. Thus, the field $\mathcal{E}_{\text{ee}}(\mathbf{r})$ may be written as a sum of its Hartree $\mathcal{E}_H(\mathbf{r})$ and Pauli-Coulomb $\mathcal{E}_{xc}(\mathbf{r})$ components:

$$\mathcal{E}_{\text{ee}}(\mathbf{r}) = \mathcal{E}_H(\mathbf{r}) + \mathcal{E}_{xc}(\mathbf{r}), \quad (24)$$

where

$$\mathcal{E}_H(\mathbf{r}) = \int \frac{\rho(\mathbf{r}')(\mathbf{r} - \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|^3} d\mathbf{r}', \quad (25)$$

and

$$\mathcal{E}_{xc}(\mathbf{r}) = \int \frac{\rho_{xc}(\mathbf{r}\mathbf{r}')(\mathbf{r} - \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|^3} d\mathbf{r}'. \quad (26)$$

Note that in traditional quantum mechanics, it is not possible to further split the Fermi-Coulomb hole into its Fermi $\rho_x(\mathbf{r}\mathbf{r}')$ and Coulomb $\rho_c(\mathbf{r}\mathbf{r}')$ components. In other words, it is not possible to separate the correlations due to the Pauli principle and Coulomb repulsion. This separation will be accomplished in Sect. V via QDFT.

The kinetic field $\mathbf{Z}(\mathbf{r})$ is defined in terms of the kinetic ‘force’ $\mathbf{z}(\mathbf{r})$ and the density $\rho(\mathbf{r})$ as

$$\mathbf{Z}(\mathbf{r}) = \frac{\mathbf{z}(\mathbf{r})}{\rho(\mathbf{r})}. \quad (27)$$

The kinetic ‘force’ is obtained from its *nonlocal* (dynamic) quantal source, the single-particle density matrix $\gamma(\mathbf{r}\mathbf{r}')$ as follows:

$$z_\alpha(\mathbf{r}) = 2 \sum_{\beta} \nabla_{\beta} t_{\alpha\beta}(\mathbf{r}; \gamma), \quad (28)$$

where the second-rank kinetic energy tensor $t_{\alpha\beta}(\mathbf{r}; \gamma)$ in Cartesian coordinates is

$$t_{\alpha\beta}(\mathbf{r}; \gamma) = \frac{1}{4} \left[\frac{\partial^2}{\partial r'_{\alpha} \partial r''_{\beta}} + \frac{\partial^2}{\partial r'_{\beta} \partial r''_{\alpha}} \right] \gamma(\mathbf{r}'\mathbf{r}'') \Big|_{\mathbf{r}'=\mathbf{r}''=\mathbf{r}}. \quad (29)$$

The quantal source $\gamma(\mathbf{r}\mathbf{r}')$ is the expectation value $\gamma(\mathbf{r}\mathbf{r}') = \langle \Psi(\mathbf{X}) | \hat{\gamma}(\mathbf{r}\mathbf{r}') | \Psi(\mathbf{X}) \rangle$, with the complex density matrix operator $\hat{\gamma}(\mathbf{r}\mathbf{r}')$ being

$$\hat{\gamma}(\mathbf{r}\mathbf{r}') = \hat{A} + i\hat{B}, \quad (30)$$

$$\hat{A} = \frac{1}{2} \sum_k [\delta(\mathbf{r}_k - \mathbf{r}) T_k(\mathbf{a}) + \delta(\mathbf{r}_k - \mathbf{r}') T_k(-\mathbf{a})], \quad (31)$$

$$\hat{B} = -\frac{i}{2} \sum_k [\delta(\mathbf{r}_k - \mathbf{r}) T_k(\mathbf{a}) - \delta(\mathbf{r}_k - \mathbf{r}') T_k(-\mathbf{a})], \quad (32)$$

with $T_k(\mathbf{a})$ a translation operator such that $T_k(\mathbf{a})\psi(\dots \mathbf{r}_k, \dots) = \psi(\dots \mathbf{r}_k + \mathbf{a}, \dots)$ and $\mathbf{a} = \mathbf{r}' - \mathbf{r}$. The operators \hat{A} and \hat{B} are each Hermitian.

The differential density field $\mathcal{D}(\mathbf{r})$ whose quantal source is the *local* electron density $\rho(\mathbf{r})$, is defined in terms of the corresponding ‘force’ $d(\mathbf{r})$ and density $\rho(\mathbf{r})$ as

$$\mathcal{D}(\mathbf{r}) = \frac{d(\mathbf{r})}{\rho(\mathbf{r})}, \quad (33)$$

where

$$d(\mathbf{r}) = -\frac{1}{4} \nabla \nabla^2 \rho(\mathbf{r}). \quad (34)$$

The magnetic field contribution $\mathcal{I}_m(\mathbf{r})$ to the internal field in terms of the ‘force’ $\mathbf{i}_m(\mathbf{r})$ and the density $\rho(\mathbf{r})$ is

$$\mathcal{I}_m(\mathbf{r}) = \frac{\mathbf{i}_m(\mathbf{r})}{\rho(\mathbf{r})}, \quad (35)$$

where

$$i_{m,\alpha}(\mathbf{r}) = \sum_{\beta} \nabla_{\beta} I_{\alpha\beta}(\mathbf{r}), \quad (36)$$

and the second-rank tensor $I_{\alpha\beta}(\mathbf{r})$ in Cartesian coordinates is

$$I_{\alpha\beta}(\mathbf{r}) = [j_{\alpha}(\mathbf{r})A_{\beta}(\mathbf{r}) + j_{\beta}(\mathbf{r})A_{\alpha}(\mathbf{r})] - \rho(\mathbf{r})A_{\alpha}(\mathbf{r})A_{\beta}(\mathbf{r}), \quad (37)$$

with $\mathbf{j}(\mathbf{r})$ the quantal source of the field.

The individual components of the internal field $\mathcal{F}^{\text{int}}(\mathbf{r})$ are in general not conservative. However, as shown below, their sum taken together with the Lorentz field is conservative. Under conditions of certain symmetry, the individual components can each be separately conservative.

C. ‘Quantal Newtonian’ First Law

The equation of motion or QNFL is satisfied by *each electron* of the physical system defined by the many-electron SP equation of Eq. (6). The law states that the sum of the external $\mathcal{F}^{\text{ext}}(\mathbf{r})$ and internal $\mathcal{F}^{\text{int}}(\mathbf{r})$ fields experienced by each electron vanishes:

$$\mathcal{F}^{\text{ext}}(\mathbf{r}) + \mathcal{F}^{\text{int}}(\mathbf{r}) = 0. \quad (38)$$

The law is derived employing the continuity condition $\nabla \cdot \mathbf{j}(\mathbf{r}) = 0$.

Thus, the quantal source-field perspective of the SP theory is consistent with Schrödinger’s [2] insight that satisfaction of this condition is the explanation of the lack of radiation in a stationary state. The QNFL is valid for arbitrary state. It is also gauge invariant.

D. Total Energy and Components

The terms of the total energy E – the canonical kinetic T , the electron-interaction E_{ee} , and its Hartree E_H and Pauli-Coulomb E_{xc} components, – can each be expressed in integral virial form in terms of the corresponding fields $\mathcal{Z}(\mathbf{r})$, $\mathcal{E}_{\text{ee}}(\mathbf{r})$, $\mathcal{E}_H(\mathbf{r})$, $\mathcal{E}_{xc}(\mathbf{r})$. With the exception of $\mathcal{E}_H(\mathbf{r})$ which is conservative, these expressions are valid irrespective of

whether the fields are conservative. Thus,

$$T = -\frac{1}{2} \int \rho(\mathbf{r}) \mathbf{r} \cdot \mathbf{Z}(\mathbf{r}) d\mathbf{r}, \quad (39)$$

$$E_{ee} = \int \rho(\mathbf{r}) \mathbf{r} \cdot \mathbf{E}_{ee}(\mathbf{r}) d\mathbf{r}, \quad (40)$$

$$E_H = \int \rho(\mathbf{r}) \mathbf{r} \cdot \mathbf{E}_H(\mathbf{r}) d\mathbf{r}, \quad (41)$$

$$E_{xc} = \int \rho(\mathbf{r}) \mathbf{r} \cdot \mathbf{E}_{xc}(\mathbf{r}) d\mathbf{r}. \quad (42)$$

The contribution of the conservative external electrostatic field $\mathbf{E}(\mathbf{r}) = -\nabla v(\mathbf{r})$ to the energy E_{es} can be written directly in terms of the potential $v(\mathbf{r})$ as

$$E_{es} = \int \rho(\mathbf{r}) v(\mathbf{r}) d\mathbf{r}. \quad (43)$$

Note that $v(\mathbf{r})$ is *path-independent*. The energy can also be written in integral virial form, but the coefficient of the expression depends upon the degree of the homogeneous function $v(\mathbf{r})$. Hence, for the Coulombic potential for which the degree is -1 , the expression is

$$E_{es} = \int \rho(\mathbf{r}) \mathbf{r} \cdot \mathbf{E}(\mathbf{r}) d\mathbf{r}. \quad (44)$$

For the magnetic field contribution to the energy, *i.e.* the contribution of the Lorentz $\mathbf{L}(\mathbf{r})$ and internal magnetic $\mathcal{I}_m(\mathbf{r})$ field components, define the field

$$\mathcal{M}(\mathbf{r}) = -[\mathbf{L}(\mathbf{r}) + \mathcal{I}_m(\mathbf{r})]. \quad (45)$$

If the field $\mathcal{M}(\mathbf{r})$ is conservative, *i.e.* $\nabla \times \mathcal{M}(\mathbf{r}) = 0$, then one can define a magnetic scalar potential $v_m(\mathbf{r})$ as

$$\mathcal{M}(\mathbf{r}) = -\nabla v_m(\mathbf{r}). \quad (46)$$

This implies that $v_m(\mathbf{r})$ is *path-independent*. The magnetic contribution E_{mag} to the energy is then

$$E_{\text{mag}} = \int \rho(\mathbf{r}) v_m(\mathbf{r}) d\mathbf{r}. \quad (47)$$

The E_{mag} can also be written in integral virial form depending on the degree of the homogeneous function $v_m(\mathbf{r})$. If $v_m(\mathbf{r})$ is of degree 2 as for the harmonic oscillator, then

$$E_{\text{mag}} = -\frac{1}{2} \int \rho(\mathbf{r}) \mathbf{r} \cdot \mathcal{M}(\mathbf{r}) d\mathbf{r}. \quad (48)$$

In the general case when $\nabla \times \mathcal{M}(\mathbf{r}) \neq 0$, the expression is

$$E_{\text{mag}} = \int \rho(\mathbf{r}) \mathbf{r} \cdot \mathcal{M}(\mathbf{r}) d\mathbf{r}. \quad (49)$$

The total energy E may then be expressed as

$$E = T + E_{\text{ee}} + E_{\text{es}} + E_{\text{mag}} \quad (50)$$

$$= T + E_H + E_{\text{xc}} + E_{\text{es}} + E_{\text{mag}}. \quad (51)$$

It is evident from the above that the quantum-mechanical system defined via the SP equation can be alternatively described from the perspective of the individual electron in terms of the fields experienced by it.

E. Further Physical and Mathematical Insights

In addition to the above new perspective, further understandings of the SP system may be gleaned from the QNFL. These are as follows:

(i) In summing the QNFL over all the electrons, the contribution of the internal field $\mathcal{F}^{\text{int}}(\mathbf{r})$ vanishes, leading to Ehrenfest's [21] theorem for a stationary state: $\int \rho(\mathbf{r}) \mathcal{F}^{\text{ext}}(\mathbf{r}) d\mathbf{r} = 0$.

(ii) The external scalar potential $v(\mathbf{r})$ is the binding potential, and it is path-independent. The QNFL, however, provides a deeper physical understanding of this potential in terms of the properties of the system. Further, it affords an interpretation of the potential in the rigorous classical sense. It follows from the QNFL of Eq. (38) that *the potential $v(\mathbf{r})$ is the work done to move an electron from some reference point at infinity to its position at \mathbf{r} in the force of a conservative field $\mathcal{F}(\mathbf{r})$* :

$$v(\mathbf{r}) = \int_{\infty}^{\mathbf{r}} \mathcal{F}(\mathbf{r}') \cdot d\boldsymbol{\ell}', \quad (52)$$

where $\mathcal{F}(\mathbf{r}) = \mathcal{F}^{\text{int}}(\mathbf{r}) - \mathcal{L}(\mathbf{r}) = \mathcal{E}_{\text{ee}}(\mathbf{r}) - \mathcal{Z}(\mathbf{r}) - \mathcal{D}(\mathbf{r}) - \mathcal{I}_m(\mathbf{r}) - \mathcal{L}(\mathbf{r})$. As the field $\mathcal{F}(\mathbf{r})$ is conservative, the $\nabla \times \mathcal{F}(\mathbf{r}) = 0$. Hence, the work done is *path-independent*, and therefore $v(\mathbf{r})$ constitutes a potential energy. It is reiterated that the QNFL is valid for arbitrary state. Hence, the potential function $v(\mathbf{r})$ as expressed in Eq. (52) remains the *same* irrespective of the state of the system.

(*iii*) In the Hamiltonian of Eq. (5), the potential energy function $v(\mathbf{r})$ binding the electrons is assumed analytically known. It could be Coulombic ($-Ze^2/r$), harmonic ($\frac{1}{2}kr^2$), screened-Coulomb Yukawa ($-Ze^2\exp(-\lambda r/r)$, etc. The QNFL written as in Eq. (52) then shows that this analytical function $v(\mathbf{r})$ depends on all the components of the internal field $\mathcal{F}^{\text{int}}(\mathbf{r})$ of the system and the Lorentz field $\mathcal{L}(\mathbf{r})$. Thus, the potential $v(\mathbf{r})$ is inherently related to and constructed via the properties of the system. Further, if the various internal fields are separately conservative, then the function $v(\mathbf{r})$ is comprised of a sum of constituent functions, each representative of a property of the system, with each being the work done in the corresponding field. (See the example of Sect. III.)

(*iv*) Provided the sum of the Lorentz $\mathcal{L}(\mathbf{r})$ and internal magnetic $\mathcal{I}_m(\mathbf{r})$ fields is conservative, it is then possible to define a scalar potential $v_m(\mathbf{r})$ representative of all the magnetic effects of the system. This potential is the work done in the sum of the fields $\mathcal{L}(\mathbf{r})$ and $\mathcal{I}_m(\mathbf{r})$. This work done is path-independent.

(*v*) The QNFL also provides a deeper mathematical understanding of the potential $v(\mathbf{r})$. As the components of the conservative field $\mathcal{F}(\mathbf{r})$ of Eq. (52) are obtained from quantal sources that are expectation values of Hermitian operators taken with respect to the wave function Ψ , the field $\mathcal{F}(\mathbf{r})$ is a functional of Ψ , *i.e.* $\mathcal{F}(\mathbf{r}) = \mathcal{F}[\Psi](\mathbf{r})$. This functional is *exactly known* since the individual component fields are explicitly defined. This in turn means that the scalar potential energy $v(\mathbf{r})$ as defined by Eq. (52) is an *exactly known* functional of the wave function Ψ : $v(\mathbf{r}) = v[\Psi](\mathbf{r})$. We emphasize that this functional dependence is valid for *arbitrary* state. (That the external potential $v(\mathbf{r})$ is a functional of the *ground state* wave function Ψ_g was originally proved by Hohenberg and Kohn [16] for the case when the only external field present was the electrostatic binding field $\mathcal{E}(\mathbf{r})$. The explicit functional dependence of $v(\mathbf{r})$ on Ψ_g was, however, not given.)

III. ‘QUANTAL NEWTONIAN’ FIRST LAW FOR THE 2^3S STATE OF A QUANTUM DOT IN A MAGNETIC FIELD

In this section we demonstrate the satisfaction of the QNFL by application to the triplet 2^3S state of a 2-electron (with spin) 2-dimensional quantum dot in a magnetic field. A detailed study of this state of the quantum dot exhibiting in particular the significance of the magnetization current density $\mathbf{j}_m(\mathbf{r})$ is given in [13]. The results presented are taken

from this work.

In quantum dots, the motion of the electrons is confined to 2 dimensions in a quantum well in a thin layer of semiconductor such as GaAs which is sandwiched between two layers of another semiconductor AlGaAs. The 2-dimensional motion of the electrons is restricted by an electrostatic field that can be varied. This motion can be further constrained by a magnetic field perpendicular to the plane of motion. As the ‘artificial atom’ is in a semiconductor, the free electron mass m must be replaced by the band effective mass m^* , and the electron- interaction modified by the dielectric constant ϵ . For GaAs the effective mass is $m^* = 0.067m$, and $\epsilon = 12.4$. Finally, the binding potential $v(\mathbf{r})$ of the electrons has been established via both theory and experiment to be harmonic [10] so that $v(\mathbf{r}) = \frac{1}{2}m^*\omega_0^2r^2$.

The SP equation for the quantum dot in a magnetic field is the following:

$$\left[\frac{1}{2m^*} \sum_{k=1}^2 (\hat{\mathbf{p}}_k + \frac{e}{c} \mathbf{A}(\mathbf{r}_k))^2 + g^* \mu_B \sum_{k=1}^2 \mathbf{B}(\mathbf{r}_k) \cdot \mathbf{s}_k + \frac{1}{2\epsilon} \sum_{k,\ell=1}^2 \frac{1}{|\mathbf{r}_k - \mathbf{r}_\ell|} + \frac{1}{2} m^* \sum_{k=1}^2 \omega_0^2 r_k^2 \right] \Psi(\mathbf{X}) = E \Psi(\mathbf{X}), \quad (53)$$

with $k_0 = \omega_0^2$ the binding force constant, $\mu_B = e\hbar/2m$ the Bohr magneton, and g^* the corresponding gyromagnetic ratio.

(Effective atomic units are employed: $e^2/\epsilon = \hbar = m^* = c = 1$. The effective Bohr radius is $a_0^* = a_0(m/m^*)$, where m is the free electron mass. The effective energy unit is $(a.u.)^* = (a.u.)(m^*/m\epsilon^2)$.)

In the symmetric gauge $\mathbf{A}(\mathbf{r}) = \frac{1}{2} \mathbf{B}(\mathbf{r}) \times \mathbf{r}$, and $\mathbf{B}(\mathbf{r}) = \mathcal{B} \hat{\mathbf{i}}_z$, the wave function $\Psi(\mathbf{x}_1 \mathbf{x}_2) = \psi(\mathbf{r}_1 \mathbf{r}_2) \chi(\sigma_1 \sigma_2)$, with the *exact* closed-form analytical expression for the spatial part being

$$\psi(\mathbf{r}_1 \mathbf{r}_2) = N e^{im\theta} e^{-\Omega(r_1^2 + r_2^2)/2} [u + c_2 u^2 + c_3 u^3 + c_4 u^4], \quad (54)$$

where the normalization constant $N = 0.022466$; the angular momentum quantum number $m = 1$; the effective oscillator frequency $\Omega^2 = \omega_0^2 + \omega_L^2$ or effective force constant $k_{\text{eff}} = \Omega^2 = 0.072217$ where $\omega_L = \mathcal{B}/2 = 0.1$ is the Larmor frequency; the relative coordinate vector $\mathbf{u} = \mathbf{r}_2 - \mathbf{r}_1$; the angle θ is that of the vector \mathbf{u} ; the coefficients $c_2 = \frac{1}{3}$; $c_3 = -0.059108$; $c_4 = -0.015884$. Note that the spatial part $\psi(\mathbf{r}_1 \mathbf{r}_2)$ is *antisymmetric* in an interchange of \mathbf{r}_1 and \mathbf{r}_2 due to the phase factor $e^{im\theta}$.

For the triplet 2^3S state of the quantum dot, the QNFL of Eq. (38) written in

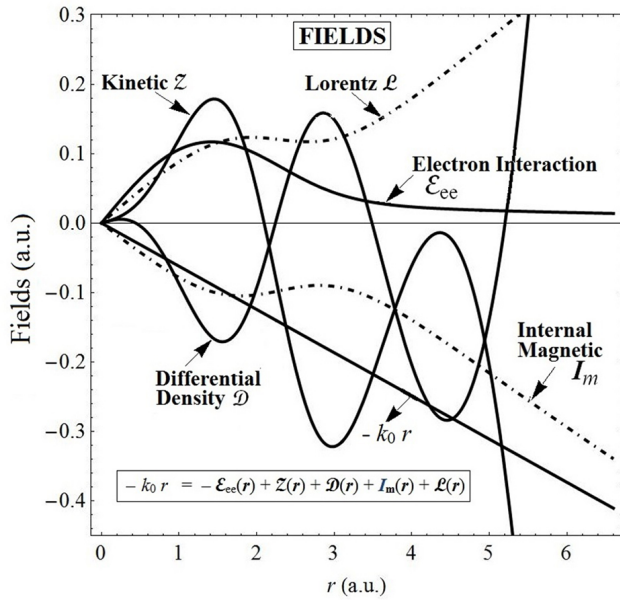


FIG. 1: The fields experienced by each electron: electron-interaction $\mathcal{E}_{ee}(r)$; kinetic $\mathcal{Z}(r)$; differential density $\mathcal{D}(r)$; Lorentz $\mathcal{L}(r)$; and internal magnetic $\mathcal{I}_m(r)$. The fields $\mathcal{L}(r)$ and $\mathcal{I}_m(r)$ are plotted for a value of the Larmor frequency of $\omega_L = 0.1$. Also plotted is the function $-k_0 r$.

terms of the individual fields as defined in Sect. II, and the binding force constant k_0 , is

$$-k_0 r = -\mathcal{E}_{ee}(\mathbf{r}) + \mathcal{Z}(\mathbf{r}) + \mathcal{D}(\mathbf{r}) + \mathcal{L}(\mathbf{r}) + \mathcal{I}_m(\mathbf{r}). \quad (55)$$

The individual fields: the electron-interaction $\mathcal{E}_{ee}(\mathbf{r})$; kinetic $\mathcal{Z}(\mathbf{r})$; differential density $\mathcal{D}(\mathbf{r})$; Lorentz $\mathcal{L}(\mathbf{r})$; and internal magnetic $\mathcal{I}_m(\mathbf{r})$, are plotted in Fig. 1. The figure shows that on summing the various disparate fields, one obtains the linear function corresponding to the gradient of the binding potential. This demonstrates the satisfaction of the QNFL.

From the fields shown in Fig. 1, the individual components of the total energy E are determined via the definitions of Sect. II D. Thus, in effective atomic units (*a.u.*)*, $T = 0.615577$, $E_H = 0.755497$, $E_{xc} = -0.501339$, $E_{ee} = 0.254158$, $E_{es} + E_{mag} = 0.742657$. The energy E of this triplet state of the quantum dot is then $E = 1.612391$ (*a.u.*)*. The first Ionization Potential is -1.343659 (*a.u.*)*. These values are the same as those obtained as the expectations of the corresponding operators taken with respect to the wave function $\Psi(\mathbf{x}_1\mathbf{x}_2)$.

IV. GENERALIZATION OF THE SCHRÖDINGER-PAULI EQUATION

Another consequence of the QNFL is the generalization of the SP equation. This generalized form exhibits its intrinsic self-consistent nature. As shown in Sect. II, the scalar binding potential $v(\mathbf{r})$ is an exactly known functional of the wave function Ψ . Substituting the functional $v[\Psi](\mathbf{r})$ into the SP equation Eq. (6), the equation can be written as

$$\left[\frac{1}{2} \sum_k (\hat{\mathbf{p}}_k + \frac{1}{c} \mathbf{A}(\mathbf{r}_k))^2 + \frac{1}{c} \sum_k \mathcal{B}(\mathbf{r}_k) \cdot \mathbf{s}_k + \frac{1}{2} \sum'_{k,\ell} \frac{1}{|\mathbf{r}_k - \mathbf{r}_\ell|} + \sum_k v[\Psi](\mathbf{r}_k) \right] \Psi(\mathbf{X}) = E[\Psi] \Psi(\mathbf{X}), \quad (56)$$

or, on employing Eq. (52), as

$$\left[\frac{1}{2} \sum_k (\hat{\mathbf{p}}_k + \frac{1}{c} \mathbf{A}(\mathbf{r}_k))^2 + \frac{1}{c} \sum_k \mathcal{B}(\mathbf{r}_k) \cdot \mathbf{s}_k + \frac{1}{2} \sum'_{k,\ell} \frac{1}{|\mathbf{r}_k - \mathbf{r}_\ell|} + \sum_k \int_{\infty}^{\mathbf{r}_k} \mathcal{F}[\Psi](\mathbf{r}) \cdot d\boldsymbol{\ell} \right] \Psi(\mathbf{X}) = E[\Psi] \Psi(\mathbf{X}). \quad (57)$$

Thus, the Hamiltonian is a functional of the wave function $\Psi : \hat{H} = \hat{H}[\Psi]$, and the SP equation can then be written in its generalized form as in Eq. (7). The Hamiltonian

functional $\hat{H}[\Psi]$ is *exactly known*. It is valid for *arbitrary state*. It is also *universal* in that it is applicable to any electronic system defined by this Hamiltonian.

The generalized form of the SP equation makes evident that the eigenfunctions Ψ and eigenvalues E may be obtained self-consistently. Given a Hamiltonian \hat{H} with a *known* binding potential $v(\mathbf{r})$, the procedure is the following. One begins with an approximate input wave function Ψ^{in} to first determine the corresponding quantal sources and fields, and the approximate potential $v[\Psi^{in}](\mathbf{r})$ via Eq. (52), and thereby the approximate Hamiltonian $\hat{H}[\Psi^{in}]$ and energy $E[\Psi^{in}]$. (The approximate $v[\Psi^{in}](\mathbf{r})$ differs from the known $v(\mathbf{r})$.) The SP equation $\hat{H}[\Psi^{in}]\Psi^{out} = E[\Psi^{out}]\Psi^{out}$ is then solved to obtain the output approximation $\{\Psi^{out}, E[\Psi^{out}]\}$ to the wave function and energy. This output wave function Ψ^{out} then becomes the *new input* wave function, and leads to the next approximate potential $v[\Psi^{out}](\mathbf{r})$ which is also *not* the known $v(\mathbf{r})$. Then the new $\{\Psi^{out}, E[\Psi^{out}]\}$ is obtained. The above procedure is continued till the $\Psi^{in} = \Psi^{out}$ and $E[\Psi^{out}] = E[\Psi^{in}]$. When this is achieved then $v[\Psi^{out} = \Psi^{in}](\mathbf{r})$ will correspond to the known $v(\mathbf{r})$. In this manner, the exact $\{\Psi(\mathbf{X}), E\}$ can be determined. Why the above self-consistent procedure is possible comes from the understanding of what the functional $v[\Psi](\mathbf{r})$ means. *The meaning of the functional $v[\Psi](\mathbf{r})$ is that for each new Ψ , one obtains a new $v(\mathbf{r})$, and therefore the Hamiltonian functional $\hat{H}[\Psi]$ changes with each new iterative Ψ .* This is what allows for the self-consistent procedure. As in all self-consistent calculations, the input approximate wave function must be physically reasonable for the self-consistent procedure to converge.

V. QUANTAL DENSITY FUNCTIONAL THEORY

In this section the system of electrons described by the SP Hamiltonian of Eq. (5) is mapped via quantal density functional theory (QDFT) [6, 7] to one of noninteracting fermions possessing the same $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$. The additional constraints on the model system are that it also possess the same number N of fermions, and the same total orbital \mathbf{L} and spin \mathbf{S} angular momentum. It is assumed that the model fermions are subject to the same electrostatic $\mathcal{E}(\mathbf{r}) = -\nabla v(\mathbf{r})/e$ and magnetostatic $\mathcal{B}(\mathbf{r}) = \nabla \times \mathbf{A}(\mathbf{r})$ fields as the electrons of the interacting system. It is further assumed that such a model system can exist.

The key to the mapping from the interacting to the noninteracting fermion model system is to determine the *local* electron-interaction potential $v_{ee}(\mathbf{r})$ in which the many-

body effects are incorporated. This potential then generates the single-particle orbitals $\phi_i(\mathbf{x})$ of the Slater determinant $\Phi\{\phi_i(\mathbf{x})\}$ that lead to the same electronic and physical current density. For the QDFT model system with the constraints as described above, the only electron correlations that must be *explicitly* accounted for in $v_{ee}(\mathbf{r})$ are those due to the Pauli exclusion principle and Coulomb repulsion, and Correlation-Kinetic effects [22].

It is reiterated that a principal purpose of the mapping to the model system is to determine the Correlation-Kinetic energy T_c , and to separate the Pauli-Coulomb E_{xc} (quantum-mechanical exchange-correlation) energy into its Pauli E_x (exchange) and Coulomb E_c (correlation) energy components.

Consider a system of N *noninteracting* fermions possessing the same potential energies $\{v, \mathbf{A}\}$ as that of the interacting electrons. The SP Hamiltonian \hat{H}_s of the model fermions (the S system) is (see also Eq. (16))

$$\hat{H}_s = \hat{T} + \frac{1}{c} \int \hat{\mathbf{j}}_s(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) d\mathbf{r} - \frac{1}{2c^2} \int \hat{\rho}(\mathbf{r}) A^2(\mathbf{r}) d\mathbf{r} + \hat{V}_s, \quad (58)$$

where the local potential operator \hat{V}_s is

$$\hat{V}_s = \sum_k v_s(\mathbf{r}_k) = \sum_k \left[v(\mathbf{r}_k) + v_{ee}(\mathbf{r}_k) \right], \quad (59)$$

and $v_{ee}(\mathbf{r})$ the *local* electron-interaction potential in which all the many-body effects are incorporated. As the configuration of the model fermions is as yet unspecified, the S system current density operator $\hat{\mathbf{j}}_s(\mathbf{r})$ is

$$\hat{\mathbf{j}}_s(\mathbf{r}) = \hat{\mathbf{j}}_p(\mathbf{r}) + \hat{\mathbf{j}}_d(\mathbf{r}) + \hat{\mathbf{j}}_{m,s}(\mathbf{r}), \quad (60)$$

with the paramagnetic $\hat{\mathbf{j}}_p(\mathbf{r})$ and diamagnetic $\hat{\mathbf{j}}_d(\mathbf{r})$ current density operators defined as in Eqs. (10) and (11). The magnetization current density operator $\hat{\mathbf{j}}_{m,s}(\mathbf{r}) = -c \nabla \times \hat{\mathbf{m}}_s(\mathbf{r})$ with the magnetization density operator $\hat{\mathbf{m}}_s(\mathbf{r}) = (-\frac{1}{c}) \sum_k \mathbf{s}_{k,s} \delta(\mathbf{r}_k - \mathbf{r})$, and $\mathbf{s}_{k,s}$ the spin vector of the k -th model fermion.

For *arbitrary state* of the interacting system, the mapping to the model system is to be such that it possesses the same basic variables $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ and satisfies the same constraints on N, \mathbf{L} , and \mathbf{S} . With the orbital angular momentum \mathbf{L} being the same, the equivalence of the spin \mathbf{S} angular momentum requires that $\mathbf{s}_{k,s} = \mathbf{s}_k$. (This means that the configuration of the model fermions is either the same as that of the interacting electrons, or a different configuration but one possessing the same \mathbf{L} and \mathbf{S} . Thus, for example, it is

possible to map an interacting two-electron system in an excited singlet state to a S system in its ground state.) The equivalence of the spin vectors implies that $\hat{\mathbf{m}}_s(\mathbf{r}) = \mathbf{m}(\mathbf{r})$ so that $\hat{\mathbf{j}}_{m,s}(\mathbf{r}) = \hat{\mathbf{j}}_m(\mathbf{r})$. It follows that the operator $\hat{\mathbf{j}}_s(\mathbf{r}) = \hat{\mathbf{j}}(\mathbf{r})$. Hence, the S system Hamiltonian \hat{H}_s of Eq. (58) may be written as

$$\hat{H}_s = \hat{T} + \frac{1}{c} \int \hat{\mathbf{j}}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) d\mathbf{r} - \frac{1}{2c^2} \int \hat{\rho}(\mathbf{r}) A^2(\mathbf{r}) + \hat{V}_s. \quad (61)$$

The corresponding local effective potential differential equation for the orbitals $\phi_i(\mathbf{x})$ of the Slater determinant wave function $\Phi\{\phi_k\}$ of the model fermions (assuming additionally that $c = 1$) is

$$\left[\frac{1}{2} (\hat{\mathbf{p}} + \mathbf{A}(\mathbf{r}))^2 + \mathbf{B} \cdot \mathbf{s} + v(\mathbf{r}) + v_{ee}(\mathbf{r}) \right] \phi_k(\mathbf{x}) = \epsilon_k \phi_k(\mathbf{x}); \quad k = 1, \dots, N. \quad (62)$$

The S system properties of the density $\rho_s(\mathbf{r})$, Dirac density matrix $\gamma_s(\mathbf{r}\mathbf{r}')$, pair-correlation density $g_s(\mathbf{r}\mathbf{r}')$, and the current density $\mathbf{j}_s(\mathbf{r})$ are then, respectively, the expectation values of the corresponding Hermitian operators taken with respect to the Slater determinant wave function $\Phi\{\phi_i\}$. Thus $\rho_s(\mathbf{r}) = \langle \Phi\{\phi_k\} | \hat{\rho}(\mathbf{r}) | \Phi\{\phi_k\} \rangle = \sum_{\sigma,k} \phi_k^*(\mathbf{r}\sigma) \phi_k(\mathbf{r}\sigma)$; $\gamma_s(\mathbf{r}\mathbf{r}') = \langle \Phi\{\phi_k\} | \hat{\gamma}(\mathbf{r}\mathbf{r}') | \Phi\{\phi_k\} \rangle = \sum_{\sigma,k} \phi_k^*(\mathbf{r}\sigma) \phi_k(\mathbf{r}'\sigma)$; $g_s(\mathbf{r}\mathbf{r}') = \frac{1}{\rho_s(\mathbf{r})} \langle \Phi\{\phi_k\} | \hat{P}(\mathbf{r}\mathbf{r}') | \Phi\{\phi_k\} \rangle$; $\mathbf{j}_s(\mathbf{r}) = \langle \Phi\{\phi_k\} | \hat{\mathbf{j}}(\mathbf{r}) | \Phi\{\phi_k\} \rangle$. Note that $g_s(\mathbf{r}\mathbf{r}') = \rho_s(\mathbf{r}') + \rho_x(\mathbf{r}\mathbf{r}')$, where the Fermi hole is defined as $\rho_x(\mathbf{r}\mathbf{r}') = -|\gamma_s(\mathbf{r}\mathbf{r}')|^2 / 2\rho_s(\mathbf{r})$.

With the requirement that the S system density $\rho_s(\mathbf{r})$ and current density $\mathbf{j}_s(\mathbf{r})$ are the same as $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ of the interacting system, the QNFL as satisfied by each model fermion is then

$$\mathcal{F}^{\text{ext}}(\mathbf{r}) + \mathcal{F}_s^{\text{int}}(\mathbf{r}) = 0. \quad (63)$$

The law is derived (see Appendix A) employing the continuity condition $\nabla \cdot \mathbf{j}(\mathbf{r}) = 0$. As the potentials $\{v, \mathbf{A}\}$ and the densities $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ of the S system are the same as those of the interacting system, the external field $\mathcal{F}^{\text{ext}}(\mathbf{r})$ experienced by the model fermions is the same as for the electrons (see Eq. (17)). The internal field $\mathcal{F}_s^{\text{int}}(\mathbf{r})$ of these fermions is obtained as

$$\mathcal{F}_s^{\text{int}}(\mathbf{r}) = -\nabla v_{ee}(\mathbf{r}) - \mathcal{Z}_s(\mathbf{r}) - \mathcal{D}(\mathbf{r}) - \mathcal{I}_m(\mathbf{r}), \quad (64)$$

where $\mathcal{Z}_s(\mathbf{r}), \mathcal{D}(\mathbf{r}), \mathcal{I}_m(\mathbf{r})$ are the corresponding kinetic, differential density, and internal magnetic fields. The S system kinetic field $\mathcal{Z}_s(\mathbf{r})$ is defined in a manner similar to the kinetic field $\mathcal{Z}(\mathbf{r})$ of the interacting system (see Eq. (30)):

$$\mathcal{Z}_s(\mathbf{r}) = \frac{z_s(\mathbf{r})}{\rho(\mathbf{r})}, \quad (65)$$

where the kinetic ‘force’ $\mathbf{z}_s(\mathbf{r})$ is obtained from its *nonlocal* quantal source, the Dirac density matrix $\gamma_s(\mathbf{r}\mathbf{r}')$ (defined earlier) as

$$z_{s,\alpha}(\mathbf{r}) = 2 \sum_{\beta} \nabla_{\beta} t_{s,\alpha\beta}(\mathbf{r}; \gamma_s), \quad (66)$$

where the second rank tensor $t_{s,\alpha\beta}(\mathbf{r}; \gamma_s)$ is

$$t_{s,\alpha\beta}(\mathbf{r}; \gamma_s) = \frac{1}{4} \left[\frac{\partial^2}{\partial r'_{\alpha} \partial r''_{\beta}} + \frac{\partial^2}{\partial r'_{\beta} \partial r''_{\alpha}} \right] \gamma_s(\mathbf{r}'\mathbf{r}'') \Big|_{\mathbf{r}'=\mathbf{r}''=\mathbf{r}}. \quad (67)$$

The fields $\mathbf{D}(\mathbf{r})$ and $\mathbf{I}_m(\mathbf{r})$ are defined as for the interacting system. As the densities $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ of the interacting and S systems are the same, so are these corresponding fields (see Eqs. (33) and (35)).

Equating the ‘Quantal Newtonian’ first laws for the interacting and model systems (Eqs. (38) and (63)) then leads to the definition of the *local* electron-interaction potential $v_{ee}(\mathbf{r})$ of the S system differential equation (Eq. (62)). *The potential $v_{ee}(\mathbf{r})$ is the work done to move the model fermion from some reference point at infinity to its position at \mathbf{r} in the force of a conservative effective field $\mathcal{F}^{\text{eff}}(\mathbf{r})$:*

$$v_{ee}(\mathbf{r}) = - \int_{\infty}^{\mathbf{r}} \mathcal{F}^{\text{eff}}(\mathbf{r}') \cdot d\mathbf{l}', \quad (68)$$

where

$$\mathcal{F}^{\text{eff}}(\mathbf{r}) = \mathcal{E}_{ee}(\mathbf{r}) + \mathcal{Z}_{t_c}(\mathbf{r}), \quad (69)$$

with the electron-interaction field $\mathcal{E}_{ee}(\mathbf{r})$ given by Eq. (21), and the correlation-kinetic field $\mathcal{Z}_{t_c}(\mathbf{r})$ defined as

$$\mathcal{Z}_{t_c}(\mathbf{r}) = \mathcal{Z}_s(\mathbf{r}) - \mathcal{Z}(\mathbf{r}). \quad (70)$$

Since the $\nabla \times \mathcal{F}^{\text{eff}}(\mathbf{r})$ vanishes, the potential $v_{ee}(\mathbf{r})$ is *path-independent*. Further, in the self-consistent determination of $v_{ee}(\mathbf{r})$, it follows from Eq. (69) that the only correlations that must be accounted for are those due to the Pauli exclusion principle, Coulomb repulsion, and correlation-kinetic effects.

The total energy E of the interacting system can be expressed in terms of the S system properties. Splitting the kinetic energy T into its noninteracting T_s and correlation-kinetic T_c components, the energy $E = \langle \Psi(\mathbf{X}) | \hat{H} | \Psi(\mathbf{X}) \rangle$ (assuming $c = 1$) may be written as (see Eq. (16))

$$E = T_s + \int \rho(\mathbf{r})v(\mathbf{r})d\mathbf{r} + \int \mathbf{j}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r})d\mathbf{r} - \frac{1}{2} \int \rho(\mathbf{r})A^2(\mathbf{r})d\mathbf{r} + E_{ee} + T_c. \quad (71)$$

By multiplying the S system differential equation Eq. (62) by $\phi_k^*(\mathbf{x})$, summing over all the model fermions, and integrating over all space, the noninteracting kinetic energy T_s is obtained as

$$T_s = \sum_k \epsilon_k - \int \rho(\mathbf{r})v(\mathbf{r})d\mathbf{r} - \int \rho(\mathbf{r})v_{ee}(\mathbf{r})d\mathbf{r} - \int \mathbf{j}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r})d\mathbf{r} + \frac{1}{2} \int \rho(\mathbf{r})A^2(\mathbf{r})d\mathbf{r}. \quad (72)$$

In substituting Eq. (72) into Eq. (71) the expression for E becomes

$$E = \sum_k \epsilon_k - \int \rho(\mathbf{r})v_{ee}(\mathbf{r})d\mathbf{r} + E_{ee} + T_c, \quad (73)$$

where

$$T_c = \frac{1}{2} \int \rho(\mathbf{r})\mathbf{r} \cdot \mathcal{Z}_{t_c}(\mathbf{r})d\mathbf{r}. \quad (74)$$

Thus, the correlation-kinetic energy T_c – the contribution of electron correlations to the kinetic energy – is explicitly defined. This is a property of the electronic system not obtainable solely by solution of the SP equation.

Finally, via the mapping to the model S system, it is possible to split the Pauli-Coulomb energy E_{xc} of Eq. (42) into its Pauli E_x and Coulomb E_c components. Defining the Coulomb hole $\rho_c(\mathbf{r}\mathbf{r}')$ as the difference between the Fermi-Coulomb $\rho_{xc}(\mathbf{r}\mathbf{r}')$ and Fermi $\rho_x(\mathbf{r}\mathbf{r}')$ hole charges where $\rho_x(\mathbf{r}\mathbf{r}')$ is determined from $\gamma_s(\mathbf{r}\mathbf{r}')$ as mentioned earlier: $\rho_c(\mathbf{r}\mathbf{r}') = \rho_{xc}(\mathbf{r}\mathbf{r}') - \rho_x(\mathbf{r}\mathbf{r}')$, the Pauli-Coulomb field $\mathcal{E}_{xc}(\mathbf{r})$ of Eq. (29) may be written as a sum of its Pauli $\mathcal{E}_x(\mathbf{r})$ and Coulomb $\mathcal{E}_c(\mathbf{r})$ components:

$$\mathcal{E}_{xc}(\mathbf{r}) = \mathcal{E}_x(\mathbf{r}) + \mathcal{E}_c(\mathbf{r}), \quad (75)$$

where

$$\mathcal{E}_x(\mathbf{r}) = \int \frac{\rho_x(\mathbf{r}\mathbf{r}')(\mathbf{r} - \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|^3}d\mathbf{r}', \quad (76)$$

and

$$\mathcal{E}_c(\mathbf{r}) = \int \frac{\rho_c(\mathbf{r}\mathbf{r}')(\mathbf{r} - \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|^3}d\mathbf{r}'. \quad (77)$$

The corresponding Pauli E_x and Coulomb E_c energies are then, respectively,

$$E_x = \int \rho(\mathbf{r})\mathbf{r} \cdot \mathcal{E}_x(\mathbf{r})d\mathbf{r}, \quad (78)$$

and

$$E_c = \int \rho(\mathbf{r})\mathbf{r} \cdot \mathcal{E}_c(\mathbf{r})d\mathbf{r}. \quad (79)$$

The total energy E of the interacting system may thus be written in terms of its components as (see Eq. (51))

$$E = T_s + E_H + E_x + E_c + T_c + E_{es} + E_{\text{mag}}. \quad (80)$$

(Note that T_s may be determined either via the field $\mathcal{Z}_s(\mathbf{r})$ through its integral virial expression, or as the expectation value $\langle \Phi\{\phi_k\} | \hat{T} | \Phi\{\phi_k\} \rangle$). It is emphasized that the components of the energy E are properties of the *same* physical system.

The fact that the energy E can be so expressed in terms of these individual components shows the significance of the QDFT mapping to the model system. As such the mapping constitutes an essential complement to SP theory. The mapping to the model system possessing the same basic variables of the density and physical current density provides additional properties of the physical system.

The second principal reason for the development of a local effective potential theory such as the QDFT described above, or Kohn-Sham density functional theory, is the easier numerical solution of the corresponding single-particle differential equation. These theories, of course, lead to properties of the interacting system. The QDFT differential equation Eq. (62) is designed to deliver the interacting system density $\rho(\mathbf{r})$, and the current density $\mathbf{j}(\mathbf{r})$. From these properties may be determined all single-particle operator expectation values, the Lorentz ‘force’ and field, and the internal magnetic ‘force’ and field. The highest occupied eigenvalue of the differential equation is the negative of the ionization potential. The total energy E is in turn determined via Eq. (73).

VI. PHYSICAL INTERPRETATION OF SPIN DENSITY FUNCTIONAL THEORY

Kohn-Sham density functional theory (KS-DFT) [16], a ground state theory, also constitutes a mapping from the interacting system of electrons in an external field $\mathcal{E}(\mathbf{r}) = -\nabla v(\mathbf{r})/e$ to one of noninteracting fermions possessing the same density $\rho(\mathbf{r})$. All the many-body effects are incorporated in an electron-interaction energy functional $E_{ee}^{KS}[\rho]$ with the corresponding local potential being the functional derivative $v_{ee}(\mathbf{r}) = \delta E_{ee}^{KS}[\rho]/\delta\rho(\mathbf{r})$. A rigorous physical interpretation of the functional $E_{ee}^{KS}[\rho]$ and its derivative $v_{ee}(\mathbf{r})$ via QDFT in terms of ‘classical’ fields representative of the different electron correlations is given in [6].

The Kohn-Sham spin density functional theory (SDFT) [8–10] that we consider is

the original extension of KS-DFT to electrons with spin in the added presence of a magnetostatic field $\mathbf{B}(\mathbf{r}) = \nabla \times \mathbf{A}(\mathbf{r})$. In this section we briefly review the principal tenets of this SDFT. We then construct a *modified* SDFT, and provide a physical interpretation of the corresponding functionals and functional derivatives based on the QDFT mapping of the previous section. Finally, we show the original SDFT to be a special case of the modified version.

SDFT is a ground state theory. The interacting system Hamiltonian on which SDFT is based is an approximation to the SP Hamiltonian of Eq. (5). The approximate Hamiltonian is

$$\hat{H}^{\text{SDFT}} = \frac{1}{2} \sum_k \hat{\mathbf{p}}_k^2 + \frac{1}{c} \sum_k \mathbf{B}(\mathbf{r}_k) \cdot \mathbf{s}_k + \hat{W} + \hat{V}, \quad (81)$$

$$= \hat{T} + \int \hat{\mathbf{m}}(\mathbf{r}) \cdot \mathbf{B}(\mathbf{r}) d\mathbf{r} + \hat{W} + \hat{V}, \quad (82)$$

with the various terms defined in Sect. II. What is missing in this Hamiltonian is the field component $(\frac{1}{c})\mathbf{A}(\mathbf{r})$ of the electron momentum. (Compare with Eq. (5) of SP theory.) Hence, the interaction of the magnetic field with the orbital angular momentum is absent in this description. Note that this Hamiltonian is not derivable from first principles and is *ad hoc*. Based on this Hamiltonian, it is assumed that the basic variables are the electronic $\rho(\mathbf{r})$ and magnetization $\mathbf{m}(\mathbf{r})$ densities. Or equivalently, the basic variables are the electronic density $\rho(\mathbf{r})$ and $Q(\mathbf{r})$ which is the difference between the spin-up $\rho^\alpha(\mathbf{r})$ and spin-down $\rho^\beta(\mathbf{r})$ densities. (As noted in the Introduction, the basic variables consistent with the SP Hamiltonian of Eq. (5) are [17] the electronic $\rho(\mathbf{r})$ and the physical current $\mathbf{j}(\mathbf{r})$ densities, together with the constraints of constant electron number N , and orbital \mathbf{L} and spin \mathbf{S} angular momentum.)

The SDFT energy functional $E^{\text{SDFT}}[\rho, \mathbf{m}]$ corresponding to the Hamiltonian of Eq. (81) is then [10]

$$E^{\text{SDFT}}[\rho, \mathbf{m}] = T_s[\rho, \mathbf{m}] + \int \mathbf{m}(\mathbf{r}) \cdot \mathbf{B}(\mathbf{r}) d\mathbf{r} + \int \rho(\mathbf{r}) v(\mathbf{r}) d\mathbf{r} + E_{ee}^{\text{SDFT}}[\rho, \mathbf{m}], \quad (83)$$

where $T_s[\rho, \mathbf{m}]$ is the model system kinetic energy functional, and $E_{ee}^{\text{SDFT}}[\rho, \mathbf{m}] = E_{ee}^{\text{SDFT}}[\rho^\alpha, \rho^\beta]$ the functional in which all the many-body effects are incorporated.

The assumption that $\{\rho, \mathbf{m}\}$ are the basic variables implies that the wave function $\Psi(\mathbf{X})$ corresponding to the Hamiltonian \hat{H}^{SDFT} of Eq. (81) is a functional of $\{\rho, \mathbf{m}\}$. For

this to be the case, it must be proved that there is a one-to-one or bijective relationship between $\{\rho, \mathbf{m}\}$ and the potentials $\{v, \mathbf{A}\}$ (or equivalently $\{v, \mathbf{B}\}$). There exists no proof that the ground state $\{\rho, \mathbf{m}\}$ uniquely determine $\{v, \mathbf{A}\}$ to within a constant and gradient of a scalar function while *simultaneously* explicitly accounting for the many-to-one relationship between $\{v, \mathbf{A}\}$ and the ground state $\Psi(\mathbf{X})$.

There is a Percus-Levy-Lieb [23] (PLL) constrained-search formulation [7] for the existence of the universal functional $E_{ee}^{\text{SDFT}}[\rho, \mathbf{m}]$. But again, the proof depends on the assumption that the basic variables are $\{\rho, \mathbf{m}\}$. As has been noted [24], all PLL-type proofs require the pre-knowledge of what properties constitute the basic variables. The constrained-search is then over all antisymmetric functions that deliver those basic variables and minimize the expectation of the operators $\hat{T} + \hat{W}$. (The original PLL proof [23] of density functional theory requires the knowledge that the ground state density $\rho(\mathbf{r})$ is the basic variable. This information is provided by the Hohenberg-Kohn theorem [16], together with the constraint of constant electron number N . In a similar manner, it is only following the Pan-Sahni [17] proof that the ground state density $\rho(\mathbf{r})$ and physical current density $\mathbf{j}(\mathbf{r})$ are the basic variables that a subsequent PLL-type proof becomes viable.)

The corresponding equations of SDFT are:

$$\left[\frac{1}{2} \hat{\mathbf{p}}^2 + \mathcal{B} \cdot \mathbf{s} + v(\mathbf{r}) + v_{ee}^{\alpha, \beta}(\mathbf{r}) \right] \phi_k(\mathbf{x}) = \epsilon_k \phi_k(\mathbf{x}); \quad k = 1, \dots, N^\alpha; N^\beta, \quad (84)$$

where $v_{ee}^{\alpha, \beta}(\mathbf{r}) = \delta E_{ee}^{\text{SDFT}}[\rho^\alpha, \rho^\beta] / \delta \rho^{\alpha, \beta}(\mathbf{r})$, and $N^\alpha; N^\beta$ the number of electrons of α and β spins, respectively.

In order to explain the physical interpretation of SDFT, *i.e.* to understand the meaning of the functional $E_{ee}^{\text{SDFT}}[\rho^\alpha, \rho^\beta]$ and its functional derivatives $v_{ee}^{\alpha, \beta}(\mathbf{r})$, we first construct a *modified* SDFT designated MSDFT. (The MSDFT is a special case of the QDFT of the previous section.) The equations governing the MSDFT are a generalization of SDFT (Eqs. (83) and (84)) in that the field component of the momentum is now explicitly included. It is still assumed that the basic variables are $\{\rho, \mathbf{m}\}$. Thus, the energy functional $E^{\text{MSDFT}}[\rho^\alpha, \rho^\beta]$ is

$$\begin{aligned} E^{\text{MSDFT}}[\rho^\alpha, \rho^\beta] = & T_s[\rho^\alpha, \rho^\beta] + \int \rho(\mathbf{r}) v(\mathbf{r}) d\mathbf{r} + \int \mathbf{j}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) d\mathbf{r} \\ & - \frac{1}{2} \int \rho(\mathbf{r}) A^2(\mathbf{r}) d\mathbf{r} + E_{ee}^{\text{MSDFT}}[\rho^\alpha, \rho^\beta], \end{aligned} \quad (85)$$

and the differential equations are

$$\left[\frac{1}{2}(\hat{\mathbf{p}} + \mathbf{A}(\mathbf{r}))^2 + \mathbf{B} \cdot \mathbf{s} + v(\mathbf{r}) + v_{ee}^{\alpha,\beta}(\mathbf{r}) \right] \phi_k(\mathbf{x}) = \epsilon_k \phi_k(\mathbf{x}); \quad k = 1, \dots, N^\alpha; N^\beta \quad (86)$$

where the modified

$$v_{ee}^{\alpha,\beta}(\mathbf{r}) = \frac{\delta E_{ee}^{\text{MSDFT}}[\rho^\alpha, \rho^\beta]}{\delta \rho^{\alpha,\beta}(\mathbf{r})}. \quad (87)$$

(Note $E_{ee}^{\text{MSDFT}}[\rho, \mathbf{m}] = E_{ee}^{\text{MSDFT}}[\rho^\alpha, \rho^\beta]$.)

Prior to providing the interpretation of the functional $E_{ee}^{\text{MSDFT}}[\rho^\alpha, \rho^\beta]$ and its functional derivative $v_{ee}^{\alpha,\beta}(\mathbf{r})$, we rewrite the energy E corresponding to the SP Hamiltonian of Eq. (5) as given by Eq. (71) in the DFT terminology. As the basic variables of the SP system are $\{\rho, \mathbf{j}\}$, the energy E is

$$\begin{aligned} E[\rho, \mathbf{j}] = & T_s[\rho, \mathbf{j}] + \int \rho(\mathbf{r})v(\mathbf{r})d\mathbf{r} + \int \mathbf{j}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r})d\mathbf{r} \\ & - \frac{1}{2} \int \rho(\mathbf{r})A^2(\mathbf{r})d\mathbf{r} + E_{ee}[\rho, \mathbf{j}], \end{aligned} \quad (88)$$

where

$$E_{ee}[\rho, \mathbf{j}] = E_{ee} + T_c. \quad (89)$$

Hence, the functional $E_{ee}[\rho, \mathbf{j}]$ is representative of electron correlations due to the Pauli principle, Coulomb repulsion and Correlation-Kinetic effects.

Observe the similarity of $E^{\text{MSDFT}}[\rho, \mathbf{m}]$ (Eq. (85)) to $E[\rho, \mathbf{j}]$ (Eq. (88)). The question that needs to be addressed is under what conditions are the basic variables $\{\rho, \mathbf{j}\}$ equivalent to $\{\rho, \mathbf{m}\}$? Recall that the physical current density $\mathbf{j}(\mathbf{r})$ is the sum of the paramagnetic $\mathbf{j}_p(\mathbf{r})$, diamagnetic $\mathbf{j}_d(\mathbf{r})$, and magnetization $\mathbf{j}_m(\mathbf{r})$ currents. If the solution $\Psi(\mathbf{X})$ to the SP equation Eq.(6) is *real*, then the paramagnetic current density $\mathbf{j}_p(\mathbf{r}) = 0$. Hence, $\mathbf{j}(\mathbf{r}) = \mathbf{j}_d(\mathbf{r}) + \mathbf{j}_m(\mathbf{r})$. But $\mathbf{j}_d(\mathbf{r}) \sim \rho(\mathbf{r})$ (see Eq. (11)), and $\mathbf{j}_m(\mathbf{r}) \sim \mathbf{m}(\mathbf{r})$ (see Eq. (12)). Thus, the physical current density $\mathbf{j}(\mathbf{r}) \sim \mathbf{m}(\mathbf{r})$. Under such a condition, the basic variables are then $\{\rho, \mathbf{m}\}$. As the SP Hamiltonian is real, the *ground state* wave function $\Psi(\mathbf{X})$ can always be chosen to be real. (For many systems in their *ground state*, the solutions $\Psi(\mathbf{X})$ of the SP equation are real. For example, the ground state of the quantum dot considered in Sect. III is real.) The corresponding Slater determinant $\Phi\{\phi_k\}$ wave function of the model system can also be chosen to be real. Further, as the Kohn-Sham mapping is from a ground state of the interacting system to a ground state of the model system of the same configuration, the constraints of constant N , \mathbf{L} and \mathbf{S} are automatically satisfied.

It is evident then that

$$E_{ee}^{\text{MSDFT}}[\rho, \mathbf{m}] = E_{ee}[\rho, \mathbf{j}] \Big|_{\mathbf{j}_p=0}, \quad (90)$$

so that the functional $E_{ee}^{\text{MSDFT}}[\rho, \mathbf{m}]$ accounts for the Pauli and Coulomb correlations as well as the contributions of these correlations to the kinetic energy. Finally, the *functional derivatives* $v_{ee}^{\alpha, \beta}(\mathbf{r})$ of Eq. (87) represent the work done to move a fermion from a reference point at infinity to its position at \mathbf{r} in the force of the conservative effective fields $\mathcal{F}^{\alpha, \beta}(\mathbf{r})$:

$$v_{ee}^{\alpha, \beta}(\mathbf{r}) = - \int_{\infty}^{\mathbf{r}} \mathcal{F}^{\alpha, \beta}(\mathbf{r}') \cdot d\boldsymbol{\ell}', \quad (91)$$

where $\mathcal{F}^{\alpha, \beta}(\mathbf{r}) = \mathcal{F}^{\text{eff}}(\mathbf{r})$ of Eq. (69) but employing the densities $\rho^{\alpha, \beta}(\mathbf{r})$ and $\mathbf{j}^{\alpha, \beta}(\mathbf{r}) = \mathbf{j}(\mathbf{r}) \Big|_{\mathbf{j}_p=0; \rho^{\alpha, \beta}}$

The above is the physical interpretation of MSDFT. As SDFT constitutes a special case of MSDFT, the interpretation of $E_{ee}^{\text{SDFT}}[\rho^{\alpha}, \rho^{\beta}]$ and $\delta E_{ee}^{\text{SDFT}}[\rho^{\alpha}, \rho^{\beta}]/\delta \rho^{\alpha, \beta}(\mathbf{r})$ in terms of fields and electron correlations follows. This understanding of the physics should aid in the construction of approximate energy functionals with SDFT.

VII. SUMMARY OF NEW UNDERSTANDINGS AND FUTURE WORK

There are two distinct facets to the paper. The significance of each component and what is learned is summarized below. Following the summary, the extension of these ideas to the temporal case, and relativistic theory are proposed.

(a) The present understanding of stationary-state Schrödinger-Pauli (SP) theory of electrons is the traditional one, *viz.* that of the statistical Copenhagen interpretation of quantum mechanics. The quantum state of the system is then described by the *N-electron* wave function. Observable properties of the system are obtained as expectation values of Hermitian operators taken with respect to this wave function. The present work, which lies within this framework, *complements* this understanding. The new insight is that it is possible to describe the *many-electron* SP theory from the perspective of the *individual* electron in the sea of electrons via its equation of motion or ‘Quantal Newtonian’ first law (QNFL).

The law is rigorously derived from the SP equation. The law is in terms of ‘classical’ fields and quantal sources. The statement of the law is that each electron experiences

both an external and internal field, the sum of which vanishes. The quantal sources are expectations of Hermitian operators taken with respect to the wave function. Hence, the new perspective hews to the probabilistic interpretation of quantum mechanics. As in classical physics, the corresponding fields experienced by each electron pervade all space, and are tangible in the classical sense. In that context, the fields are determinate. Thus, the new perspective is simultaneously probabilistic and deterministic.

The QNFL leads to a deeper *physical* understanding of the quantum system. It also leads to a *mathematical* insight into the SP differential equation.

(i) The new physical understanding is that in addition to the *external* binding electrostatic and Lorentz fields, each electron also experiences internal fields. These *internal* fields are representative of properties of the system: Electron-correlations due to the Pauli principle and Coulomb repulsion; Kinetic effects; the electron density; and an internal magnetic field component. That such fields exist within the SP description of a many-electron quantum system was previously unknown.

These ideas and the role played by the electron spin moment are explicated by application to the first excited triplet 2^3S state of a quantum dot in a magnetic field.

(ii) The mathematical insight that the QNFL leads to is that the Hamiltonian operator \hat{H} of the system is an *exactly known* and *universal* functional of the wave function Ψ , *i.e.* $\hat{H} = \hat{H}[\Psi]$. (By universal is meant that the functional is the *same* for *all* electronic systems, *i.e.* for *arbitrary* binding potential.) This then means that the SP equation is an intrinsically *self-consistent* one. Thus, given the Hamiltonian of a system, the corresponding eigenfunctions $\Psi(\mathbf{X})$ and eigenenergies E may be determined in a self-consistent manner. Whilst one readily accepts that the Hartree-Fock theory or Local Effective Potential theories such as Kohn-Sham theory are self-consistent, we now learn that the fundamental equation from which these theories are derived too is self-consistent.

(b) The second component of the paper is on Local Effective Potential Theory (LEPT). The interacting system of electrons with spin angular momentum as defined by the SP equation is mapped via Quantal Density Functional Theory (QDFT) to one of noninteracting fermions possessing the same density $\rho(\mathbf{r})$ and physical current density $\mathbf{j}(\mathbf{r})$, and the same electron number N , orbital \mathbf{L} and spin \mathbf{S} angular momentum. The description of the model system is once again in terms of ‘classical’ fields, quantal sources, and the corresponding QNFL. Such a description is *distinct* from any existing in the literature.

The attributes of such a mapping are the following:

(i) The model system of noninteracting fermions allows for the determination of properties of the interacting system that are not obtainable solely by solution of the SP equation. For example, it is possible to determine the Correlation-Kinetic energy, *i.e.* the contribution to the kinetic energy due to electron correlations arising from the Pauli principle and Coulomb repulsion. Further, it is possible to separate the correlations due to the Pauli principle and those due to Coulomb correlations. Finally, the highest occupied eigenvalue of the model system is the negative of the Ionization Potential or Electron Affinity. And this whilst *ensuring* that these properties correspond to the interacting system $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$. (This differs from the traditional quantum chemistry methodology.) Thus, the QDFT model system also constitutes a *complement* to SP theory.

(ii) The model system of noninteracting fermions possessing the same $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r})\}$ can also be thought of as an alternative description of the interacting system. All the many-body effects are now incorporated in a local effective potential, and this potential is *explicitly defined* in terms of a *conservative* effective field within QDFT. As the corresponding differential equation has only multiplicative potential energy operators, it is more amenable to numerical solution.

(iii) The principal other LEPT incorporating electron spin, is Spin density

functional theory (SDFT). This theory is in terms of the energy functionals of the spin-polarized density, with the corresponding potentials defined as their functional derivatives. These are strictly mathematical definitions. As the definitions within QDFT are in terms of fields, it is then possible to provide a mathematically rigorous physical description of the energy functionals and functional derivatives of SDFT. This would then allow for physically based improvement of the existing approximate SDFT energy functionals. It could also be of use in other methodologies that attempt to incorporate electron correlations into Kohn-Sham theory [25], and those that combine wave function and density functional theories [26–28]. The physical interpretation is thus of value.

A generalization of the stationary-state SP theory as described here would be the extension to the temporal case. Hence, in addition to the external binding electrostatic field $\mathcal{E}(\mathbf{r}) = -\nabla v(\mathbf{r})/e$, the electrons would be subject to a time-dependent electromagnetic field: $\mathbf{E}(\mathbf{y}) = -\nabla\phi(\mathbf{y}) - (1/c)\partial\mathbf{A}(\mathbf{y})/\partial t$; $\mathcal{B}(\mathbf{y}) = \nabla \times \mathbf{A}(\mathbf{y})$, with $\mathbf{y} = (\mathbf{r}, t)$. This would then lead to the time-dependent equation of motion for each electron or equivalently the ‘Quantal Newtonian’ second law. The law would then give rise to further insights into time-dependent SP theory as in the present work. One could go beyond the Born-Oppenheimer approximation by assuming a time-dependent binding potential $\mathcal{E}(\mathbf{y}) = -\nabla v(\mathbf{y})/e$.

The stationary-state SP equation can be derived as the non-relativistic limit of the time-independent Dirac equation. For a particle of charge q and mass m in a static electromagnetic field defined by the potentials $\{v, \mathbf{A}\}$, the Dirac equation for the 2-component spinors $\psi(\mathbf{r})$ and $\eta(\mathbf{r})$ which make up the four-component spinor $\chi(\mathbf{r})$, the solution to the Dirac equation, reduces to the coupled equations :

$$E\psi(\mathbf{r}) = c\hat{\mathbf{p}}_{\text{phys}} \cdot \boldsymbol{\sigma}\eta(\mathbf{r}) + (qv + mc^2)\psi(\mathbf{r}), \quad (92)$$

$$E\eta(\mathbf{r}) = c\hat{\mathbf{p}}_{\text{phys}} \cdot \boldsymbol{\sigma}\psi(\mathbf{r}) + (qv - mc^2)\eta(\mathbf{r}). \quad (93)$$

In the non-relativistic limit, the small component $\eta(\mathbf{r})$ can be written in terms of the large component $\psi(\mathbf{r})$. Substituting this expression for $\eta(\mathbf{r})$ into Eq. (83) then leads to the SP equation for the particle. It is first proposed to further generalize the ideas presented in this paper to the above Dirac equation, and then to extend them to the many particle case.

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APPENDIX A: DERIVATION OF THE ‘QUANTAL NEWTONIAN’ FIRST LAW FOR SCHRÖDINGER-PAULI THEORY

Consider first a system of N *spinless electrons* in an external electrostatic $\boldsymbol{\mathcal{E}}(\mathbf{r}) = -\nabla v(\mathbf{r})/e$ and magnetostatic $\boldsymbol{\mathcal{B}}(\mathbf{r}) = \nabla \times \mathbf{A}(\mathbf{r})$ field. The SP theory equation for the system is

$$\hat{H}_{\text{spinless}}\Psi = E\Psi, \quad (\text{A1})$$

where the Hamiltonian $\hat{H}_{\text{spinless}}$ (charge of electron $-e$, $|e| = \hbar = m = 1$)

$$\hat{H}_{\text{spinless}} = \hat{T}_A + \hat{W} + \hat{V}, \quad (\text{A2})$$

with

$$\hat{T}_A = \frac{1}{2} \sum_k \left(\hat{\mathbf{p}}_k + \frac{1}{c} \mathbf{A}(\mathbf{r}_k) \right)^2; \quad \hat{W} = \frac{1}{2} \sum'_{k,\ell} \frac{1}{|\mathbf{r}_k - \mathbf{r}_\ell|}; \quad \hat{V} = \sum_k v(\mathbf{r}_k), \quad (\text{A3})$$

the physical kinetic, electron-interaction, and scalar potential operators, respectively.

A method [6, 29, 30] for deriving the QNFL in general is to write the wave function as $\Psi = \Psi^R + i\Psi^I$, where Ψ^R and Ψ^I are the real and imaginary parts, substitute it into the corresponding differential equation, perform the various derivatives, employ the equation of continuity, and after considerable algebra [29], arrive at the law. The law for the spin-less electron is the vanishing of the sum of an external $\mathcal{F}^{\text{ext}}(\mathbf{r})$ and internal $\mathcal{F}^{\text{int}}(\mathbf{r})$ fields.

$$\mathcal{F}^{\text{ext}}(\mathbf{r}) + \mathcal{F}^{\text{int}}(\mathbf{r}) = 0. \quad (\text{A4})$$

where

$$\mathcal{F}^{\text{ext}}(\mathbf{r}) = \boldsymbol{\mathcal{E}}(\mathbf{r}) - \boldsymbol{\mathcal{L}}(\mathbf{r}), \quad (\text{A5})$$

$$\mathcal{F}^{\text{int}}(\mathbf{r}) = \boldsymbol{\mathcal{E}}_{\text{ee}}(\mathbf{r}) - \boldsymbol{\mathcal{D}}(\mathbf{r}) - \boldsymbol{\mathcal{Z}}(\mathbf{r}) - \boldsymbol{\mathcal{I}}_m(\mathbf{r}). \quad (\text{A6})$$

The various fields in Eqs. (A4) - (A6) have the same nomenclature and definitions in terms of their respective quantal sources as given in the text. There is, however, a fundamental

difference between the law for spinless electrons Eq. (A4) and that for electrons with spin Eq. (38). This occurs in the Lorentz $\mathcal{L}(\mathbf{r})$ and internal magnetic $\mathcal{I}_m(\mathbf{r})$ field components. The quantal source of these fields – the physical current density $\mathbf{j}_{\text{spinless}}(\mathbf{r})$ – is, in this case, a sum of the paramagnetic $\mathbf{j}_p(\mathbf{r})$ and diamagnetic $\mathbf{j}_d(\mathbf{r})$ components.

For electrons with spin, (the SP equation), one could employ the same methodology as described above to arrive at the corresponding QNFL of Eq. (38). Instead of providing this derivation, the law can be more easily derived via comparison by writing the Hamiltonian $\hat{H}_{\text{spinless}}$ in terms of the density $\hat{\rho}(\mathbf{r})$ and current density $\hat{\mathbf{j}}_{\text{spinless}}(\mathbf{r})$ operators. Thus,

$$\hat{H}_{\text{spinless}} = \hat{T} + \frac{1}{c} \int \hat{\mathbf{j}}_{\text{spinless}}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) d\mathbf{r} - \frac{1}{2c^2} \int \hat{\rho}(\mathbf{r}) A^2(\mathbf{r}) d\mathbf{r} + \hat{W} + \hat{V}, \quad (\text{A7})$$

with \hat{T} the canonical kinetic energy and $\hat{\mathbf{j}}_{\text{spinless}}(\mathbf{r}) = \hat{\mathbf{j}}_p + \hat{\mathbf{j}}_d(\mathbf{r})$ the physical current density operator. However, Eq. (A7) is of the same form as the Hamiltonian \hat{H} of the SP equation Eq. (16). The only difference between these two equations is that in the latter, the current density operator $\hat{\mathbf{j}}_{\text{spin}}(\mathbf{r}) = \hat{\mathbf{j}}_p + \hat{\mathbf{j}}_d(\mathbf{r}) + \hat{\mathbf{j}}_m(\mathbf{r})$. Thus, the resulting QNFL of Eq. (38) is also of the same form as Eq. (A4) but with the added contribution of the magnetization current density $\mathbf{j}_m(\mathbf{r})$ in the Lorentz $\mathcal{L}(\mathbf{r})$ and internal magnetic $\mathcal{I}_m(\mathbf{r})$ field components. Thus, the QNFL of SP theory is derived.

The QNFL of Eq. (63) for the model noninteracting fermionic system possessing the same $\{\rho(\mathbf{r}), \mathbf{j}(\mathbf{r}), N, \mathbf{L}, \mathbf{S}\}$ as that of the interacting SP system can be derived by writing the single-particle orbitals $\phi_j(\mathbf{r}) = \phi_j^R(\mathbf{r}) + i\phi_j^I(\mathbf{r})$, where $\phi_j^R(\mathbf{r})$ and $\phi_j^I(\mathbf{r})$ are the real and imaginary parts, substituting in the differential equation Eq. (62), and employing the continuity condition.

It can also be obtained by recognizing that the Schrödinger theory and SP theory Hamiltonians \hat{H}_s of the model noninteracting fermionic system (See Eq. (58)) are of the same form. The difference between the two lies in the fact that in addition to the paramagnetic $\mathbf{j}_p(\mathbf{r})$ and diamagnetic $\mathbf{j}_d(\mathbf{r})$ components, there is the presence of the magnetization current density $\mathbf{j}_{m,s}(\mathbf{r})$ in the physical current density $\mathbf{j}(\mathbf{r})$, and thus in the Hamiltonian \hat{H}_s , of the latter. Of course the corresponding differential equations, their solutions $\phi_j(\mathbf{r})$ and the

resulting fields of the two model systems differ. But the QNFL form is the same.

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