U(1)×SU(2) gauge invariance made simple for density functional approximations

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A semirelativistic density-functional theory that includes spin-orbit couplings and Zeeman fields on equal footing with the electromagnetic potentials, is an appealing framework to develop a unified first-principles computational approach for noncollinear magnetism, spintronics, orbitronics, and topological states. The basic variables of this theory include the paramagnetic current and the spin-current density, besides the particle and the spin density, and the corresponding exchange-correlation (xc) energy functional is invariant under local U(1) × SU(2) gauge transformations. The xc-energy functional must be approximated to enable practical applications, but, contrary to the case of the standard density functional theory, finding simple approximations suited to deal with realistic atomistic inhomogeneities has been a long-standing challenge. Here we propose a way out of this impasse by showing that approximate gauge-invariant functionals can be easily generated from existing approximate functionals of ordinary density-functional theory by applying a simple *minimal substitution* on the kinetic energy density, which controls the short-range behavior of the exchange hole. Our proposal opens the way to the construction of approximate, yet nonempirical functionals, which do not assume weak inhomogeneity and therefore may have a wide range of applicability in atomic, molecular, and condensed matter physics.

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I. INTRODUCTION

Density-functional methods are the most widely used approach to efficiently compute the electronic structure of atoms, molecules, and solids. Based on the Hohenberg-Kohn theorem [1], the electronic ground-state energy of interacting electrons is computed via the solution of the Schrödinger equation for fictitious noninteracting electrons—the so-called Kohn-Sham (KS) electrons [2]. At the heart of density-functional theory (DFT) lies the idea that the exchange-correlation (xc) energy, i.e., the energy due to the electron-electron interactions beyond the classical Hartree energy, can be approximated by a *universal* functional of appropriate local densities. In its original incarnation, DFT considered only an external scalar potential coupled to the charge density, which characterizes the interacting system. This means that the universal functional for the xc energy could be written as a functional of the charge density alone. However, as soon as additional couplings, e.g., the Zeeman term or the coupling to an external vector potential, are present, the universality of the xc-energy functional is lost, unless additional densities are included as fundamental variables. This observation led over the years to the creation of *multivariate* DFTs, such as spin-DFT (SDFT) [3] for including the Zeeman coupling and current-DFT (CDFT) [4] for orbital magnetism. In these theories, the spin density and the paramagnetic current density are included as basic variables, respectively. For the description of two-dimensional heterostructures [5–7] or topological insulators [8–10], spin-orbit coupling (SOC) plays a crucial role [11]. Since SOC is naturally described in terms of spin-dependent vector potentials, its density-functional treatment requires the additional inclusion of spin current densities as basic variables for a universal xc-energy functional. The corresponding extension of DFT has been dubbed spin-current-DFT (SCDFT) [12,13].

Upgrading DFT to include additional variables not only leads to universal functionals for systems with SOCs and in strong magnetic fields, but also brings forth an important physical concept, namely, gauge symmetry [14], which places strong constraints on the admissible dependence of the xc-energy functional on the basic variables and offers guidance in the construction of approximate functionals. In CDFT, for example, the xc-energy functional is invariant under *local* U(1) gauge transformations. Similarly, for spin-orbit coupled systems, the xc-energy functional is also invariant under *local* SU(2) gauge transformations (we will come back to this point in the next section extensively). In order to guarantee the U(1) invariance of the theory, Vignale and Rasolt argued that the xc energy should not be expressed in terms of the paramagnetic current, which is gauge dependent, but in terms of the gauge invariant vorticity [4]: thus, they arrived at the first universal local density approximation for electrons in a magnetic field. Subsequent experimentation showed that the U(1)-invariant vorticity is not well suited to deal with strongly inhomogeneous systems, such as atoms [15]. Later, Abedinpour, Vignale, and Tokatly proposed the generalized U(1) × SU(2) covariant vorticity as the fundamental variable for constructing gauge-invariant approximations to the xc-energy functional in SCDFT [16]. Its definition depends on an *arbitrary* choice of a "linking path" in physical space (cf. Ref. [16] for details)—an arbitrariness that, however, disappears in the limit of slowly varying densities. To the best of our knowledge, no experience has yet been gained on the practical use of the SU(2)-covariant vorticity. In any case, this quantity only emerges naturally when analyzing systems with *minor* inhomogeneities, such as an almost-uniform electron gas, and it may therefore suffer from the same shortcomings as its U(1) counterpart, when applied to atomistic systems.

One lesson we learn from these examples is that the choice of the gauge-invariant "building blocks" of the DFT is a nontrivial and important task, which requires much ingenuity

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as well as extensive experimentation on realistic systems. For example, the development of CDFT and, to a lesser extent, of SDFT, has been hampered for many years by the lack of functionals suitable to work with strong inhomogeneities. In the case of CDFT, the difficulty arose from the vorticity being too closely tied to the uniform free electron gas model. In the case of SDFT, as applied to magnetic systems with noncollinear spins, it is well known that a straightforward generalization of functionals derived in the collinear SDFT framework [17] only accounts for longitudinal fluctuations of the spin magnetization. Various attempts have been made recently [18–20] to include a dependence on transverse fluctuation of the spin magnetization within SDFT, but none of them, for different reasons, has been proven fully satisfactory.

In this paper we propose a way out of the impasse, following a suggestion by Tao and Perdew [21] who noticed that functionals of the meta-generalized-gradient-approximation (MGGA) family, such as the TPSS and similar [22], which use the kinetic energy density $\tau(\mathbf{r})$ as a basic variable, could be made current dependent by enforcing U(1) gauge invariance through the *minimal* substitution

$$\tau \to \tilde{\tau} = \tau - \frac{\mathbf{j}\cdot\mathbf{j}}{2n}, \tag{1}$$

where $\mathbf{j}$ is the *paramagnetic* current density. Earlier evidence of the relevance of their suggestion can be found in ideas by Dobson [23] and, particularly, Becke [24], whose current-dependent functional, based on a careful study of the short-range behavior of the exchange hole (x hole), greatly improved the description of degenerate ground states in open shell atoms [25]. Follow-up works extended these ideas and demonstrated their usefulness in applications [26–28].

Along similar lines, we show that DFT-MGGA forms can be readily upgraded to SCDFT-MGGA forms (i.e., they can be made spin dependent and spin-current dependent) by enforcing the U(1) × SU(2) gauge invariance through *minimal* substitutions to be performed on the curvature of the exchange hole. These substitutions implicate new quantities such as the "spin-kinetic energy" (defined below), the spin currents, *and* the spin density combined with its gradients. Additionally, a trivial modification has to account for an extra dependence of the extended on-top exchange hole on the squared modulus of the magnetization (and *no* other extra combinations). In this manner, successful approximations of ordinary DFT can be readily turned into approximations for SCDFT and noncollinear SDFT. Our proposal opens the way to the construction of approximate, yet nonempirical functionals, which do not assume weak inhomogeneity and should therefore have a wide range of applicability in atomic, molecular, and condensed matter physics.

This paper is organized as follows. In Sec. II we review the theoretical background of U(1) × SU(2) gauge invariance in SCDFT and take the opportunity to remark that the xc fields—generated, as usual, through functional derivatives of the xc-energy functional with respect to the basic densities—can exert nontrivial torques on the spin density and paramagnetic spin currents. In Sec III we derive the generalized short-range behavior of exchange-only quantities, which allows us to extract very useful U(1) × SU(2) gauge-invariant quantities for the construction of functional approximations in SCDFT and, therefore, we point out the aforementioned minimal substitutions. In Sec. IV we give examples of new approximate functionals of the MGGA form. Technical details concerning the gauge transformations of the various fundamental quantities are reported in the Appendix.

II. THEORETICAL BACKGROUND: GAUGE INVARIANCE AND XC TORQUES

We start by introducing our notational conventions: In the following we will encounter "spatial" vectors, such as the position $\mathbf{r}$ and the usual (Abelian) vector potential $\mathbf{A}$, which will be set in boldface. Components of spatial vectors are denoted by Greek subscripts, e.g., A_μ are the spatial components of the vector potential. Furthermore, we denote vectors in "spin space", such as the spin magnetization $\vec{s}$, by using an arrow. The components in spin space are denoted by Latin superscripts, e.g., s^a. We will also encounter "mixed tensors", such as the (paramagnetic) spin currents and the non-Abelian vector potentials. These are quantities that can be regarded as vectors with an additional index, i.e., "spatial vectors" carrying a spin index (e.g., $\mathbf{A}^a$) or vectors in spin space carrying a spatial index (e.g., $\vec{A}_\mu$). Formally, non-Abelian vector potentials are useful to represent spin-orbit couplings [14,29] and, in the DFT parlance, spin currents are their conjugate densities [12,13].

SCDFT is concerned with the calculation of the ground-state energy and densities of the semirelativistic many-electron Pauli Hamiltonian [30]. In comparison to the SDFT Hamiltonian, which contains only a scalar potential $v(\mathbf{r})$ and a Zeeman magnetic field $B^a(\mathbf{r})$, the SCDFT also includes an Abelian vector potential $\mathbf{A}(\mathbf{r})$ and a non-Abelian vector potential $\mathbf{A}^a(\mathbf{r})$ [31]:

$$\hat{H} = \frac{1}{2}\int d^3r\,\hat{\Psi}^\dagger(\mathbf{r})\left[-i\nabla + \frac{1}{c}\mathbf{A}(\mathbf{r}) + \frac{\mu_B}{2c}\sigma^a\mathbf{A}^a(\mathbf{r})\right]^2\hat{\Psi}(\mathbf{r}) + \hat{W} + \int d^3r\,\hat{n}(\mathbf{r})v(\mathbf{r}) + \mu_B\int d^3r\,\hat{s}^a(\mathbf{r})B^a(\mathbf{r}). \tag{2}$$

In Eq. (2) $\mu_B = 1/2c$ is the Bohr magneton, we employ Einstein's convention to sum over repeated indices ($a = x,y,z$), and a multiplication by a 2×2 identity matrix is implied for the terms which are diagonal in spin space, i.e., $-i\nabla + \frac{1}{c}\mathbf{A}(\mathbf{r})$ and $v(\mathbf{r})$. The electron-electron interaction is given by

$$\hat{W} = \frac{1}{2}\iint d^3r_1 d^3r_2\,\frac{:\hat{\Psi}^\dagger(\mathbf{r}_1)\hat{\Psi}(\mathbf{r}_1)\hat{\Psi}^\dagger(\mathbf{r}_2)\hat{\Psi}(\mathbf{r}_2):}{|\mathbf{r}_1-\mathbf{r}_2|}, \tag{3}$$

where $:\cdots:$ denotes the normal ordering of the two-component Pauli field operators $\hat{\Psi}^\dagger = (\hat{\psi}^\dagger_\uparrow\ \hat{\psi}^\dagger_\downarrow)$; finally,

$$\hat{n}(\mathbf{r}) = \hat{\Psi}^\dagger(\mathbf{r})\hat{\Psi}(\mathbf{r}) \tag{4a}$$

is the particle-density operator and

$$\hat{s}^a(\mathbf{r}) = \hat{\Psi}^\dagger(\mathbf{r})\sigma^a\hat{\Psi}(\mathbf{r}) \tag{4b}$$

is the ath component of the spin-density operator. Expanding the square and employing a partial integration [32] in Eq. (2),

one readily obtains

$$\hat{H} = \int d^3r\,\hat{\tau}(\mathbf{r}) + \hat{W} + \frac{1}{c}\int d^3r\hat{\mathbf{j}}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) + \frac{\mu_B}{2c}\int d^3r\hat{\mathbf{J}}^a(\mathbf{r})\cdot\mathbf{A}^a(\mathbf{r}) + \int d^3r\hat{n}(\mathbf{r})\tilde{v}(\mathbf{r}) + \mu_B\int d^3r\hat{s}^a(\mathbf{r})\tilde{B}^a(\mathbf{r}), \quad (5)$$

where

$$\hat{\tau} = \frac{1}{2}(\nabla\hat{\Psi}^\dagger)\cdot(\nabla\hat{\Psi}) \quad (6a)$$

is the kinetic-energy-density operator,

$$\hat{\mathbf{j}} = \frac{1}{2i}[\hat{\Psi}^\dagger\nabla\hat{\Psi} - (\nabla\hat{\Psi}^\dagger)\hat{\Psi}] \quad (6b)$$

is the paramagnetic-current operator, and

$$\hat{\mathbf{J}}^a = \frac{1}{2i}[\hat{\Psi}^\dagger\sigma^a\nabla\hat{\Psi} - (\nabla\hat{\Psi}^\dagger)\sigma^a\hat{\Psi}] \quad (6c)$$

is the paramagnetic-spin-current operator. We have also defined

$$\tilde{v} = v + \frac{1}{2c^2}\left[\mathbf{A}\cdot\mathbf{A} + \frac{\mu_B^2}{4}\mathbf{A}^a\cdot\mathbf{A}^a\right], \quad (7)$$

$$\tilde{B}^a = B^a + \frac{1}{2c^2}\mathbf{A}\cdot\mathbf{A}^a. \quad (8)$$

Given the external fields $\mathbf{A}$, $\mathbf{A}^a$, v, and B^a, the ground-state energy is the expectation value of $\hat{H}$ in the corresponding ground state $|\Psi\rangle$. The ground-state energy can be determined by means of a constrained-search minimization [33,34]:

$$E = \min_{(n,s^a,\mathbf{j},\mathbf{J}^a)}\left\{F[n,s^a,\mathbf{j},\mathbf{J}^a] + \frac{1}{c}\int d^3r\mathbf{j}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) + \frac{\mu_B}{2c}\int d^3r\mathbf{J}^a(\mathbf{r})\cdot\mathbf{A}^a(\mathbf{r}) + \int d^3rn(\mathbf{r})\tilde{v}(\mathbf{r}) + \mu_B\int d^3rs^a(\mathbf{r})\tilde{B}^a(\mathbf{r})\right\}, \quad (9)$$

with

$$F[n,s^a,\mathbf{j},\mathbf{J}^a] = \min_{|\Psi'\rangle\to(n,s^a,\mathbf{j},\mathbf{J}^a)}\langle\Psi|\hat{T}+\hat{W}|\Psi\rangle, \quad (10)$$

where the inner minimization is carried out over all the many-body wave functions yielding the prescribed set of densities and the outer minimization is carried out with respect to all N-representable densities [35]. Equation (10) defines a universal density functionals, which is the direct generalization of the universal functional in standard DFT. Assuming that the same set of densities is both interacting and noninteracting v representable, one can further decompose F,

$$F[n,s^a,\mathbf{j},\mathbf{J}^a] = T_{\rm KS}[n,s^a,\mathbf{j},\mathbf{J}^a] + E_H[n] + E_{\rm xc}[n,s^a,\mathbf{j},\mathbf{J}^a] \quad (11)$$

in terms of the Kohn-Sham (KS) kinetic energy $T_{\rm KS}[n,s^a,\mathbf{j},\mathbf{J}^a]$, the Hartree energy $E_{\rm H}[n] = \frac{1}{2}\int d^3r\int d^3r'\frac{n(\mathbf{r})n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}$, and a remainder $E_{\rm xc}[n,s^a,\mathbf{j},\mathbf{J}^a]$, which is the xc-energy functional. In this way, the problem of determining the ground-state energies is reformulated into devising practical and sufficiently accurate approximations for $E_{\rm xc}$. In order to simplify the notation, in the following we redefine the external potentials $\mu_B B^a \to B^a$ and $\frac{\mu_B}{2}\mathbf{A}^a \to \mathbf{A}^a$.

The KS equations in SCDT have the form of single-particle Pauli equations

$$\left[\frac{1}{2}\left(-i\nabla + \frac{1}{c}\boldsymbol{\mathcal{A}}_{\rm KS}\right)^2 + \mathcal{V}_{\rm KS}\right]\Phi_\mu = \varepsilon_k\Phi_k, \quad (12)$$

where

$$\boldsymbol{\mathcal{A}}_{\rm KS} = (\mathbf{A}+\mathbf{A}_{\rm xc}) + \sigma^a\left(\mathbf{A}^a + \mathbf{A}^a_{\rm xc}\right), \quad (13)$$

$$\mathcal{V}_{\rm KS} = (v + v_{\rm H} + v_{\rm xc}) + \sigma^a\left(B^a + B^a_{\rm xc}\right) + \frac{1}{2c^2}\left[(\mathbf{A}+\sigma^a\mathbf{A}^a)^2 - \boldsymbol{\mathcal{A}}^2_{\rm KS}\right], \quad (14)$$

in which $\frac{1}{c}\mathbf{A}_{\rm xc} = \frac{\delta E_{\rm xc}}{\delta\mathbf{j}(\mathbf{r})}$ is the Abelian xc-vector potential, $\frac{1}{c}\mathbf{A}^a_{\rm xc} = \frac{\delta E_{\rm xc}}{\delta\mathbf{J}^a(\mathbf{r})}$ is the ath component of the non-Abelian xc-vector potential, $B^a_{\rm xc} = \frac{\delta E_{\rm xc}}{\delta s^a(\mathbf{r})}$ is the ath component of the xc-magnetic potential (Zeeman field), $v_{\rm xc} = \frac{\delta E_{\rm xc}}{\delta n(\mathbf{r})}$ is the xc-scalar potential, and $v_H(\mathbf{r}) = \int d\mathbf{r}\frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}$ is the usual Hartree potential.

A fundamental property of $E_{\rm xc}$ is its invariance under general U(1) × SU(2) gauge transformations. We recall that a local U(1) transformation $U(\mathbf{r})$ is defined by

$$\hat{\Psi}(\mathbf{r}) \to \hat{\Psi}'(\mathbf{r}) = \exp\left[\frac{i}{c}\chi(\mathbf{r})\right]\hat{\Psi}(\mathbf{r}), \quad (15)$$

where $\chi(\mathbf{r})$ is a scalar function of the position, and a local SU(2) transformation is defined by

$$\hat{\Psi}(\mathbf{r}) \to \hat{\Psi}'(\mathbf{r}) = \exp\left[\frac{i}{c}\lambda^a(\mathbf{r})\sigma^a\right]\hat{\Psi}(\mathbf{r}) = U_{\rm S}(\mathbf{r})\hat{\Psi}(\mathbf{r}), \quad (16)$$

where $\lambda^a(\mathbf{r})$ are the components of a vector function of the position. A detailed analysis of gauge transformations in SCDFT framework is presented in Refs. [12,13]. Note that neither $F[n,s^a,\mathbf{j},\mathbf{J}^a]$ nor $T_{\rm KS}[n,s^a,\mathbf{j},\mathbf{J}^a]$ are invariant, but they both have the same transformation properties because the KS system has the same densities n, s^a, $\mathbf{j}$, and $\mathbf{J}^a$ as the interacting system. As a result

$$E_{\rm xc}[n',s'^a,\mathbf{j}',\mathbf{j}'^a] = E_{\rm xc}[n,s^a,\mathbf{j},\mathbf{J}^a], \quad (17)$$

where

$$n \to n' = n, \quad (18a)$$

$$s^a \to s'^a = R^{ab}s^b, \quad (18b)$$

$$\mathbf{j} \to \mathbf{j}' = \mathbf{j} + \frac{1}{c}n\nabla\chi - \frac{i}{2}s^a{\rm Tr}(\sigma^a U_{\rm S}^\dagger\nabla U_{\rm S}), \quad (18c)$$

and

$$\mathbf{J}^a \to \mathbf{J}'^a = R^{ab}\left[\mathbf{J}^b + \frac{1}{c}s^b\nabla\chi - \frac{i}{2}n{\rm Tr}(\sigma^b U_{\rm S}^\dagger\nabla U_{\rm S})\right]. \quad (18d)$$

R^{ab} is a 3×3 matrix describing a rotation in $\mathbb{R}^3$ around $\hat{\lambda}$—the unit vector in the direction of λ^a—by an angle $\varphi = -2|\lambda|/c$ and Tr is the trace taken with respect to spin indices. The explicit derivation of Eqs. (18) is presented in the Appendix.

The transformation of the xc fields can be readily deduced by combining the invariance of $E_{\rm xc}$—as expressed in Eq. (17)—with the transformations of the densities, as given in Eq. (18). We have

$$\mathbf{A}'_{\rm xc} = \mathbf{A}_{\rm xc}, \tag{19a}$$

$$\mathbf{A}'^{a}_{\rm xc} = R^{ab}\mathbf{A}^{b}_{\rm xc}, \tag{19b}$$

$$B'^{a}_{\rm xc} = R^{ab}\left[B^{b}_{\rm xc} + \frac{i}{2}\mathbf{A}_{\rm xc}\cdot{\rm Tr}(\sigma_b U_{\rm S}^{\dagger}\nabla U_{\rm S}) - \mathbf{A}^{b}_{\rm xc}\cdot\nabla\chi\right], \tag{19c}$$

$$v'_{\rm xc} = v_{\rm xc} - \frac{1}{c}\mathbf{A}_{\rm xc}\cdot\nabla\chi + \frac{i}{2}\mathbf{A}^{a}_{\rm xc}\cdot{\rm Tr}(\sigma^a U_{\rm S}^{\dagger}\nabla U_{\rm S}). \tag{19d}$$

Equations (19) express how the xc fields in different gauges are related. It is apparent that while $A_{{\rm xc},\mu}$ is invariant, $B^{a}_{\rm xc}$, $\mathbf{A}^{a}_{\rm xc}$, and $v_{\rm xc}$ are not invariant, but *covariant* [36]. Note that even in the case of a restricted U(1) transformation, $B^{a}_{\rm xc}$, $\mathbf{A}_{{\rm xc},\mu}$, and $v_{\rm xc}$ *do not* behave as standard Maxwellian fields and, in general, $B^{a}_{\rm xc} \neq [\nabla\times\mathbf{A}_{\rm xc}]_a$. The xc fields in SCDFT should be regarded as some effective Yang-Mills fields.

Although the xc fields are not generated by any physical field equations, they are bound to satisfy compatibility relations—having the form of conservation laws [13]—due to the invariance of $E_{\rm xc}$. Local U(1) invariance requires

$$\partial_\mu[nA_{{\rm xc},\mu} + \vec{s}\cdot\vec{A}_{{\rm xc},\mu}] = 0 \tag{20a}$$

and local SU(2) invariance implies

$$\frac{1}{2c}\partial_\mu\cdot[A_{{\rm xc},\mu}\vec{s} + n\vec{A}_{{\rm xc},\mu}] = \frac{1}{c}\vec{A}_{{\rm xc},\mu}\times\vec{j}_\mu + \vec{B}_{\rm xc}\times\vec{s}. \tag{20b}$$

In the latter expressions, we have expressed $\mathbf{A}^{a}_{\rm xc}$ as $\vec{A}_{{\rm xc},\mu}$ and, similarly, $B^{a}_{\rm xc}$ as $\vec{B}_{\rm xc}$. Of course, $\vec{A}_{{\rm xc},\mu}$ should not be confused with $A_{{\rm xc},\mu}$ (i.e, the μth component of $\mathbf{A}_{\rm xc}$). These rewritings are useful to express the torques generated by these xc fields on their conjugate densities in terms of explicit cross products. We recall that the KS system of SCDFT reproduces the interacting paramagnetic currents but may not reproduce the diamagnetic currents. Whatever the difference between the KS and interacting diamagnetic currents is, Eqs. (20a) and (20b) ensure that the stationarity conditions for the particle and spin densities are not violated: $\vec{A}_{{\rm xc},\mu}$ and $\vec{B}_{\rm xc}$ can balance any nonvanishing xc-divergence-like contribution.

There is one nontrivial case in which we can see that the solution of the KS equations in SCDFT reduces to the solution of the analogous equations in SDFT. First, note that to have $A_{{\rm xc},\mu}=0$ and $\vec{A}_{{\rm xc},\mu}=0$ the torque of $\vec{B}_{\rm xc}$ must vanish as well [see Eq. (20b)]. Yet $\vec{B}_{\rm xc}$ can be nonvanishing if it is parallel to the spin density at every point in space. If the external non-Abelian vector potential is also vanishing, we are then in a situation in which SDFT applies rigorously. Thus, in this case, we can conclude that $v_{\rm xc} = v_{\rm xc}^{\rm SDFT}$ and $\vec{B}_{\rm xc} = \vec{B}_{\rm xc}^{\rm SDFT}$.

III. SHORT-RANGE BEHAVIOR OF EXCHANGE-ONLY PAIR-CORRELATION FUNCTIONS

Importing the standard local-spin-density approximation (LSDA) in SCDFT does not allow us to fully exploit the power of SCDFT, as the LSDA only depends on the magnitude of the spin density and the particle density. Moreover, the LSDA is insensitive to strong inhomogeneities and long-range interactions. Exact exchange would be an obvious, more sophisticated choice [37], but its combination with suitable correlation functionals may require more involved computational approaches. Generalized-gradient approximations (GGAs) and, more recently, meta-GGAs [38] (MGGA)—either stand alone or combined with the Hartree-Fock method into hybrids—are the gold standard in modern DFT calculations. Here we report an analysis that points to the fact that MGGAs are ideal forms to satisfy U(1) × SU(2) gauge invariance while fulfilling other exact properties of the underlying pair-correlation functions.

We begin by reviewing known definitions about the so-called exchange hole. Assuming, as it is commonly done, that the KS states are in the form of single Slater determinants, the exchange energy can be expressed as

$$E_{\rm x} = -\frac{1}{2}\int d^3r\int d^3r'\frac{{\rm Tr}\{\Gamma(\mathbf{r},\mathbf{r}')\Gamma(\mathbf{r}',\mathbf{r})\}}{|\mathbf{r}-\mathbf{r}'|}, \tag{21}$$

where

$$\Gamma(\mathbf{r},\mathbf{r}') = \sum_{k=1}^{N}\Phi_k(\mathbf{r})\Phi_k^{\dagger}(\mathbf{r}') \tag{22}$$

is the one-body-reduced-spin density matrix obtained from the occupied spinors, which are solutions of Eq. (12). $\Gamma(\mathbf{r},\mathbf{r}')$ is a 2×2 matrix in spin space. $E_{\rm x}$ is evidently invariant under general U(1) × SU(2) gauge transformations.

$E_{\rm x}$ can be usefully expressed in terms of the x-hole function, for which a convenient definition, applicable to noncollinear spin states is

$$h_{\rm x}(\mathbf{r},\mathbf{r}') := -\frac{{\rm Tr}\{\Gamma(\mathbf{r},\mathbf{r}')\Gamma(\mathbf{r}',\mathbf{r})\}}{n(\mathbf{r})}. \tag{23}$$

In practice, the spherical average

$$h_{\rm x}(\mathbf{r},u) := \frac{1}{4\pi}\int d\Omega_{\mathbf{u}}h_{\rm x}(\mathbf{r},\mathbf{r}+\mathbf{u}) \tag{24}$$

is what really matters to the end of the calculation of the exchange energies.

In Eq. (24), the integration is carried out with respect to the solid angle $\Omega_{\mathbf{u}}$ formed by $\mathbf{r}$ and $\mathbf{u}$; $\mathbf{r}$ is the so-called reference position. Thus we rewrite

$$E_{\rm x} = 2\pi\int d^3r n(\mathbf{r})\int du\, u\, h_{\rm x}(\mathbf{r},u). \tag{25}$$

Taylor-expanding $h_{\rm x}(\mathbf{r},u)$ for small interparticle separations u, we find

$$h_{\rm x}(\mathbf{r},u) = -\frac{n(\mathbf{r})}{2}\left(1+\frac{s^a(\mathbf{r})s^a(\mathbf{r})}{n^2(\mathbf{r})}\right) - C_{\rm x}^{\rm nc}(\mathbf{r})u^2 + \cdots, \tag{26}$$

where

$$C_{h_{\rm x}}^{\rm nc} = \frac{1}{3}\left\{\left[\left(\tau - \frac{\mathbf{j}\cdot\mathbf{j}}{2n}\right) - \left(\frac{\nabla^2 n}{4} + \frac{\nabla n\cdot\nabla n}{8n}\right)\right] + \left[\left(\frac{s^a\tau^a}{n} - \frac{\mathbf{J}^a\cdot\mathbf{J}^a}{2n}\right) - \left(\frac{s^a\cdot\nabla^2 s^a}{4n} + \frac{(\nabla s^a)\cdot(\nabla s^a)}{8n}\right)\right]\right\} \tag{27}$$

is the curvature of the x hole: the superscript "nc" emphasizes that this expression differs from the analogous quantity derived earlier for spin-unpolarized or spin-polarized *but* globally collinear states. By admitting single-particle spinors, which is the natural state of affairs in noncollinear spin systems, we find terms that depend not only on the usual kinetic energy density,

$$\tau(\mathbf{r}) = \frac{1}{2}\sum_{k=1}^{N}(\partial_\mu \Phi_k^\dagger(\mathbf{r}))(\partial_\mu \Phi_k(\mathbf{r})), \tag{28}$$

but also on the *spin-kinetic energy density* defined as

$$\tau^a(\mathbf{r}) = \frac{1}{2}\sum_{k=1}^{N}(\partial_\mu \Phi_k^\dagger(\mathbf{r}))\sigma^a(\partial_\mu \Phi_k(\mathbf{r})). \tag{29}$$

Equations (26) and (27) provide the exact short-range behavior of the x-hole function in presence of particle and spin currents for noncollinear states. The gauge invariance of the expansion coefficients in Eq. (26) is obvious: a scalar function has been expanded with respect to a scalar variable. But one may also verify this property by direct inspection.

The on-top x hole [i.e., the first term on the right-hand side of Eq. (26)] provides us with an explicit indication on how the on-top x hole of an existing DFT functional should be modified to admit an extra *nonempirical* dependence on U(1) × SU(2) gauge-invariant quantities; simply, $n \to n(1 + \frac{s^a s^a}{n^2})$.

At the level of the curvature of the x hole [Eq. (27)], notice that the combination $[(\tau - \frac{\mathbf{j}\cdot\mathbf{j}}{2n}) - (\frac{\nabla^2 n}{4} + \frac{\nabla n\cdot\nabla n}{8n})]$ is already known to be a U(1) gauge-invariant quantity: the difference here is that all the quantities are evaluated on fully *noncollinear* two component spinors. Thus, this expression is *not* SU(2) gauge invariant. The contribution $[(\frac{s^a\tau^a}{n} - \frac{\mathbf{J}^a\cdot\mathbf{J}^a}{2n}) - (\frac{s^a\cdot\nabla^2 s^a}{4n} + \frac{(\nabla s^a)\cdot(\nabla s^a)}{8n})]$ is essential to get the full invariance. A detailed discussion of the transformation of each term is presented in the Appendix.

Therefore, the minimal substitution

$$\tau \to \tilde{\tau} = \left(\tau - \frac{\mathbf{j}\cdot\mathbf{j}}{2n}\right) + \left(\frac{s^a\tau^a}{n} - \frac{\mathbf{J}^a\cdot\mathbf{J}^a}{2n}\right) - \left(\frac{s^a\cdot\nabla^2 s^a}{4n} + \frac{(\nabla s^a)\cdot(\nabla s^a)}{8n}\right) \tag{30}$$

can be used to transform a DFT-MGGA form into a SCDFT-MGGA form.

In practical applications it is often desirable to eliminate the Laplacian terms, which may be difficult to evaluate numerically. To accomplish this, one substitutes the x hole, expressed in terms of $C^{\rm nc}_{h_{\rm x}}$, into the expression for the exchange energy density and performs an integration by parts to show that $C^{\rm nc}_{h_{\rm x}}$ is actually equivalent (as far as the calculation of the exchange energy is concerned) to

$$\bar{C}^{\rm nc}_{h_{\rm x}} = \frac{1}{3}\left[\left(\tau - \frac{\mathbf{j}\cdot\mathbf{j}}{2n} + \frac{\nabla n\cdot\nabla n}{8n}\right) + \left(\frac{s^a\tau^a}{n} - \frac{\mathbf{J}^a\cdot\mathbf{J}^a}{2n} + \frac{(\nabla s^a)\cdot(\nabla s^a)}{8n}\right)\right], \tag{31}$$

which no longer contains the Laplacian operator. Obviously the form of $\bar{C}^{\rm nc}_{h_{\rm x}}$ implies the minimal substitution

$$\tau \to \tilde{\tau} = \left(\tau - \frac{\mathbf{j}\cdot\mathbf{j}}{2n}\right) + \left(\frac{s^a\tau^a}{n} - \frac{\mathbf{J}^a\cdot\mathbf{J}^a}{2n}\right) + \frac{(\nabla s^a)\cdot(\nabla s^a)}{8n}. \tag{32}$$

Alternatively, functional approximations are also constructed working directly at the level of the one-body density matrix

$$Q_{\rm x}(\mathbf{r},\mathbf{r}') = {\rm Tr}\{\Gamma(\mathbf{r},\mathbf{r}')\Gamma(\mathbf{r}',\mathbf{r})\}, \tag{33}$$

in terms of which the exchange energy can be expressed as

$$E_{\rm x} = -\frac{1}{2}\int d^3r \int d^3u \frac{Q_{\rm x}(\mathbf{r}+\mathbf{u}/2,\mathbf{r}-\mathbf{u}/2)}{u} = -2\pi\int d^3r\int u\,du\,Q_{\rm x}(\mathbf{r},u), \tag{34}$$

where the vector positions are expressed with respect to the coordinates of the center of mass. Taylor-expanding $Q^{\rm nc}_{\rm x}(\mathbf{r},u)$ with respect to u, we obtain

$$Q_{\rm x}(\mathbf{r},u) = \frac{\left[n^2(\mathbf{r}) + s^a(\mathbf{r})s^a(\mathbf{r})\right]}{2} + C^{\rm nc}_{Q_{\rm x}}(\mathbf{r})u^2 + \cdots, \tag{35}$$

where

$$C^{\rm nc}_{Q_{\rm x}} = \frac{1}{3}\left[\left(n\tau - \frac{\mathbf{j}\cdot\mathbf{j}}{2} - \frac{n\nabla^2 n}{8}\right) + \left(s^a\tau^a - \frac{\mathbf{J}^a\cdot\mathbf{J}^a}{2} - \frac{s^a\nabla^2 s^a}{8}\right)\right]. \tag{36}$$

$C^{\rm nc}_{Q_{\rm x}}$ generalizes the known DFT expression to SCDFT.

Therefore, in extending a DFT-MGGA form based on the short range of $Q_{\rm x}$, we can proceed by performing two minimal substitutions: the first one, $n^2 \to (n^2 + s^a s^a)$, has to be carried out only at the level of the on-top quantities; the second one is performed at the level of τ,

$$\tau \to \tilde{\tau} = \left(\tau - \frac{\mathbf{j}\cdot\mathbf{j}}{2n}\right) + \left(s^a\tau^a - \frac{\mathbf{J}^a\cdot\mathbf{J}^a}{2}\right) - \frac{s^a\nabla^2 s^a}{8}. \tag{37}$$

Again, an intermediate integration by parts yields an alternative form of the curvature factor:

$$C^{\rm nc}_{Q_{\rm x}} \to \bar{C}^{\rm nc}_{Q_{\rm x}} = n\bar{C}^{\rm nc}_{h_{\rm x}}, \tag{38}$$

which, modulo an overall multiplication by the particle density, implies the same minimal substitution as in Eq. (32).

IV. CONSTRUCTION OF FUNCTIONALS

We conclude our analysis by constructing two new exchange-energy functionals based on existing forms and proposing them for immediate use.

First, let us consider the construction of the BR89 [39]: this was derived allowing only globally collinear spin polarization. In this approach, the x hole of the hydrogen atom is turned

into a general model by introducing two position-dependent "parameters" $p_1(\mathbf{r})$ and $p_2(\mathbf{r})$,

$$h_{\rm x}^{\rm model}(\mathbf{r},u) = -\frac{p_1(\mathbf{r})}{16\pi p_2(\mathbf{r})u}\Big\{p_1(\mathbf{r})[|p_2(\mathbf{r})-u|+1]e^{-p_1(\mathbf{r})|p_2(\mathbf{r})-u|} - p_1(\mathbf{r})[|p_2(\mathbf{r})+u|+1]e^{-p_1(\mathbf{r})|p_2(\mathbf{r})+u|}\Big\} \quad (39)$$

to be chosen in such a way to reproduce the short-range behavior of the x hole of an N-electron system.

Equations (26) and (27) allow us to readily generalize this procedure to the noncollinear current-carrying states. As a result, the $p_1(\mathbf{r})$ and $p_2(\mathbf{r})$ must be determined by solving the equations

$$p_1^3 e^{-p_1 p_2} = 4\pi n\left(1+\frac{s^a s^a}{n^2}\right), \quad (40a)$$

$$p_1^2 p_2 - 2p_1 = 12b\frac{C_{h_{\rm x}}^{\rm nc}}{n\left(1+\frac{s^a s^a}{n^2}\right)}. \quad (40b)$$

As a second example, let us consider the approximations based on a Gaussian resummation of the short-range behavior of $Q_{\rm x}(\mathbf{r},u)$. For closed-shell systems (i.e., vanishing spin polarization), Lee and Parr [40] find

$$E_{\rm x}^{\rm G} = -\pi\int d^3r n^2(\mathbf{r})\beta(\mathbf{r}). \quad (41)$$

For the sake of simplicity, here, we are not considering a more sophisticated form which would satisfy particle-number normalization for any system. In view of Eqs. (34) and (35), the extension to noncollinear spin-polarized current-caring states is readily obtained upon the substitutions

$$\beta \to -\frac{1}{2}\frac{n^2+s^a s^a}{C_{Q_{\rm x}}^{\rm nc}} \quad (42a)$$

and

$$n^2 \to (n^2+s^a s^a). \quad (42b)$$

Finally, we note that $C_{Q_{\rm x}}^{\rm nc}$ is positive for single-particle states, for states with vanishing spin currents, for the spin spirals of the uniform gas, but otherwise the question remains open.

V. CONCLUSIONS

This work opens the way to the extension of time-proven semilocal exchange-correlation energy functionals as well as to the derivation of novel approximations designed to deal with noncollinear spin structures which are subjects of great and large ongoing interest. Specifically, we have introduced nonempirical U(1) × SU(2) gauge-invariant building blocks which are, in principle, ideally suited for dealing with (static) spin fluctuation of *strongly* inhomogeneous states. Our results show how the gradients of the spin density should be combined with the spin-kinetic-energy density—an information which should be relevant even in devising semilocal forms within standard spin-DFT—and the Kohn-Sham paramagnetic (spin) currents appear as *explicit* ingredients as well. Thus, we have provided examples of extension of existing exchange-only functional forms. On passing, we have also illustrated some of the exact fundamental features of the exact exchange-correlation fields.

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APPENDIX: DERIVATION OF THE TRANSFORMATION LAWS

The expansions worked out in the main text have the advantage to spare us from the burden to explicitly deal with the transformations of the densities under general gauge transformations—this is because we could consistently deal only with scalars. Nevertheless, in this Appendix we report the derivation of the transformation laws of the considered densities and their salient combinations. This should offer thorough clarifications and further insights.

We will use the same notational convention as in the main text: Spatial indices are denoted by subscripts, spin indices by superscripts, and repeated indices are summed. Furthermore, the dependence on the position $\mathbf{r}$ is implied. We are interested in obtaining the transformation laws for the following densities:

$$n = \Phi^\dagger\Phi, \quad (A1a)$$

$$s^a = \Phi^\dagger\sigma^a\Phi, \quad (A1b)$$

$$j_\mu = \frac{1}{2i}[\Phi^\dagger(\partial_\mu\Phi) - (\partial_\mu\Phi^\dagger)\Phi], \quad (A1c)$$

$$J_\mu^a = \frac{1}{2i}[\Phi^\dagger\sigma^a(\partial_\mu\Phi) - (\partial_\mu\Phi^\dagger)\sigma^a\Phi], \quad (A1d)$$

$$\tau = \frac{1}{2}(\partial_\mu\Phi^\dagger)(\partial_\mu\Phi), \quad (A1e)$$

$$\tau^a = \frac{1}{2}(\partial_\mu\Phi^\dagger)\sigma^a(\partial_\mu\Phi). \quad (A1f)$$

The combined U(1) × SU(2) transformation is given by

$$U = \exp\left[\frac{i}{c}(\chi+\lambda^a\sigma^a)\right] = \exp\left[\frac{i}{c}\chi\right]\exp\left[\frac{i}{c}\lambda^a\sigma^a\right], \quad (A2)$$

where we use that the U(1) and SU(2) transformation commute. This means that we can investigate the U(1) and SU(2) transformation laws separately.

U(1) transformation laws: The density n and the spin magnetization s^a are trivially invariant under local U(1) transformations. It is straightforward to obtain the transformation laws for the remaining densities, i.e.,

$$n \to n, \quad (A3a)$$

$$s^a \to s^a, \quad (A3b)$$

$$j_\mu \to j_\mu + \frac{1}{c}(\partial_\mu \chi) n, \tag{A3c}$$

$$J^a_\mu \to J^a_\mu + \frac{1}{c}(\partial_\mu \chi) s^a, \tag{A3d}$$

$$\tau \to \tau + \frac{1}{c}(\partial_\mu \chi) j_\mu + \frac{1}{2c^2}(\partial_\mu \chi)(\partial_\mu \chi) n, \tag{A3e}$$

$$\tau^a \to \tau^a + \frac{1}{c}(\partial_\mu \chi) J^a_\mu + \frac{1}{2c^2}(\partial_\mu \chi)(\partial_\mu \chi) s^a. \tag{A3f}$$

It follows directly that the combinations

$$nt - \tfrac{1}{2} j_\mu j_\mu, \tag{A4a}$$

$$s^a \tau^a - \tfrac{1}{2} J^a_\mu J^a_\mu \tag{A4b}$$

are invariant under local U(1) transformations.

Infinitesimal SU(2) transformation laws: The density is trivially invariant under local SU(2) transformations. The spin magnetization, however, is not invariant. Using that

$$U_S = \exp\left[\frac{i}{c}\lambda^a \sigma^a\right] = \cos[\lambda/c] + i \sin[\lambda/c] \hat{\lambda}^a \sigma^a, \tag{A5}$$

where λ is the magnitude and $\hat{\lambda}^a$ is the unit vector in the direction of the vector λ^a. We recall

$$U_S^\dagger \sigma^a U_S = \cos[2\lambda/c]\sigma^a - \sin[2\lambda/c]\epsilon^{abc}\hat{\lambda}^b \sigma^c + (1 - \cos[2\lambda/c])\hat{\lambda}^a \hat{\lambda}^b \sigma^b = R^{ab}\sigma^b. \tag{A6}$$

The matrix R^{ab} is a 3×3 matrix describing a rotation in $\mathbb{R}^3$ around the direction $\hat{\lambda}$ by an angle $\varphi = -2\lambda/c$. It follows that the spin magnetization transforms as

$$s^a \to R^{ab} s^b. \tag{A7}$$

Before embarking on the derivation of the transformation laws for the other densities, we consider the case of *infinitesimal* transformations. This means that we can approximate

$$U_S \approx 1 + \frac{i}{c}\lambda^a \sigma^a. \tag{A8}$$

Keeping terms up to the first order in λ, we arrive at

$$n \to n, \tag{A9a}$$

$$s^a \to s^a - \frac{2\lambda}{c}\epsilon^{abc}\hat{\lambda}^b s^c, \tag{A9b}$$

$$j_\mu \to j_\mu + \frac{1}{c}(\partial_\mu \lambda^a) s^a, \tag{A9c}$$

$$J^a_\mu \to J^a_\mu - \frac{2\lambda}{c}\epsilon^{abc}\hat{\lambda}^b J^c_\mu + \frac{1}{c}(\partial_\mu \lambda^a) n, \tag{A9d}$$

$$\tau \to \tau + \frac{1}{c}(\partial_\mu \lambda^a) J^a_\mu, \tag{A9e}$$

$$\tau^a \to \tau^a - \frac{2\lambda}{c}\epsilon^{abc}\hat{\lambda}^b \tau^c + \frac{1}{c}(\partial_\mu \lambda^a) j_\mu - \frac{1}{2c}\epsilon^{abc}(\partial_\mu \lambda^b)(\partial_\mu s^c). \tag{A9f}$$

It is straightforward to verify that neither the quantity in Eq. (A4a) nor the combination in Eq. (A4b) are invariant under these transformations. Yet, the overall invariance of the x-only curvatures given in the main text can be now explicitly verified for arbitrary infinitesimal U(1) and SU(2) transformations using Eqs. (A3) and (A9). This task may be further simplified by considering the identities: $\nabla s^a \cdot \nabla s^a = \frac{\nabla^2}{2} s^a s^a - s^a \nabla^2 s^a$ and $\nabla n \cdot \nabla n = \frac{\nabla^2}{2} n^2 - n \nabla^2 n$.

Furthermore, the behavior of the basic densities under the same transformations suffices to establish the compatibility conditions (20a) and (20b) presented in the main text. This is achieved by using the fact that the xc energy is invariant under the corresponding U(1) × SU(2) transformations.

Finite SU(2) transformation laws: In principle, it is sufficient to establish invariance under infinitesimal SU(2) transformation, as an arbitrary finite SU(2) transformation can be represented as a sequence of infinitesimal transformations. However, the derivation of the transformation laws of the xc potentials requires knowledge of the explicit transformations of the basic densities under arbitrary *finite* SU(2) transformations. Moreover, we here intend to spell out the finite transformation of the spin-kinetic-energy density as well.

The difficulty in obtaining finite transformation laws is due to the fact that the generators of the SU(2) group, i.e., the Pauli matrices σ^a, do not commute. Accordingly, we have to keep in mind that

$$\partial_\mu U_S \neq \frac{i}{c}(\partial_\mu \lambda^a)\sigma^a U_S \neq \frac{i}{c} U_S (\partial_\mu \lambda^a)\sigma^a. \tag{A10}$$

Instead, using the very definition of the directional derivative, we have

$$\begin{aligned}\partial_\mu U_S &\equiv \lim_{\epsilon \to 0} \frac{1}{\epsilon}\left(\exp\left[\frac{i}{c}\lambda^a(\mathbf{r} + \epsilon \hat{k})\sigma^a\right] - \exp\left[\frac{i}{c}\lambda^a(\mathbf{r})\sigma^a\right]\right) \\ &= \lim_{\epsilon \to 0} \frac{1}{\epsilon}\left(\exp\left[\frac{i}{c}(\lambda^a + \epsilon \partial_\mu \lambda^a)\sigma^a\right] - \exp\left[\frac{i}{c}\lambda^a(\mathbf{r})\sigma^a\right]\right) \\ &= \partial_\epsilon \exp\left[\frac{i}{c}(\lambda^a + \epsilon \partial_\mu \lambda^a)\sigma^a\right]\Big|_{\epsilon=0}.\end{aligned} \tag{A11}$$

Now we can use the identity

$$\begin{aligned}\partial_\epsilon e^{\gamma(\hat{A} + \epsilon \hat{B})} &= e^{\gamma(\hat{A} + \epsilon \hat{B})} \int_0^\gamma d\gamma'\, e^{-\gamma'(\hat{A} + \epsilon \hat{B})} \hat{B} e^{\gamma'(\hat{A} + \epsilon \hat{B})} \\ &= \int_0^\gamma d\gamma'\, e^{\gamma'(\hat{A} + \epsilon \hat{B})} \hat{B} e^{-\gamma'(\hat{A} + \epsilon \hat{B})} e^{\gamma(\hat{A} + \epsilon \hat{B})},\end{aligned} \tag{A12}$$

which is readily verified by noting that both sides fulfill the same first-order differential equation in γ and vanish for $\gamma = 0$, and write

$$\begin{aligned}U_S^\dagger(\partial_\mu U_S) &= \frac{i}{c}(\partial_\mu \lambda^a) \int_0^1 d\gamma\, \exp\left[-\frac{i\gamma}{c}\lambda^b \sigma^b\right]\sigma^a \\ &\quad \times \exp\left[\frac{i\gamma}{c}\lambda^c \sigma^c\right] \\ &= \frac{i}{c}(\partial_\mu \lambda^a)\Lambda^{ab}\sigma^b,\end{aligned} \tag{A13}$$

where we have introduced

$$\Lambda^{ab} = \int_0^1 d\gamma\, R^{ab}(\gamma). \tag{A14}$$

The 3×3 matrix Λ^{ab} is the uniform average over the rotation matrices around axis $\hat{\lambda}^a$ with rotation angles $\varphi \in [0, -2\lambda/c]$. Note that Λ^{ab} *is not* a rotation matrix itself. Using the group

properties of the rotation matrices, one can easily verify the relation

$$R^{ac}\Lambda^{bc} = \Lambda^{ab}, \tag{A15}$$

which is an extremely useful identity as it shows how the rotation matrix R^{ab} acts on the matrix Λ^{ab}. Straightforward but tedious algebra allows us to write down the transformation laws for finite local SU(2) transformations in a concise form

$$n \to n, \tag{A16a}$$

$$s^a \to s'^a = R^{ab}s^b, \tag{A16b}$$

$$j_\mu \to j_\mu + \frac{1}{c}\tilde{A}^b_\mu s^b, \tag{A16c}$$

$$J^a_\mu \to R^{ab}\left[J^b_\mu + \frac{1}{c}\tilde{A}^b_\mu n\right], \tag{A16d}$$

$$\tau \to \tau + \frac{1}{c}J^a_\mu\tilde{A}^a_\mu + \frac{1}{2c^2}\tilde{A}^a_\mu\tilde{A}^a_\mu, \tag{A16e}$$

$$\tau^a \to R^{ab}\left[\tau^b + \frac{1}{c}\tilde{A}^b_\mu j_\mu + \frac{1}{2c^2}\tilde{A}^b_\mu\tilde{A}^c_\mu s^c\right] - \frac{1}{8}R^{ab}(\partial_\mu R^{cb})(\partial_\mu R^{cd})s^d + \frac{1}{4}(\partial_\mu R^{ab})(\partial_\mu s^b), \tag{A16f}$$

where we introduced $\tilde{A}^a_\mu = (\partial_\mu\lambda^b)\Lambda^{ba}$, which is the non-Abelian gauge vector potential induced by the local SU(2) transformation. Note that in the main text, following another notational choice widely adopted in the literature, we wrote $\tilde{A}^a_\mu = -\frac{ic}{2}\mathrm{Tr}(\sigma^a U_S^\dagger\nabla_\mu U_S)$.

Finally, combining the U(1) and SU(2) transformation laws for the density, spin magnetization, and the paramagnetic current and spin current—together with the invariance of E_{xc}—leads to the transformation properties of the xc potential given in the main text [Eq. (19)].

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