

Transverse Spin-Gradient Functional for Noncollinear Spin-Density-Functional Theory

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We present a novel functional for spin-density-functional theory aiming at the description of noncollinear magnetic structures. The construction of the functional employs the spin-spiral-wave state of the uniform electron gas as reference system. We show that the functional depends on transverse gradients of the spin magnetization; i.e., in contrast with the widely used local spin density approximation, the functional is sensitive to local changes of the direction of the spin magnetization. As a consequence the exchange-correlation magnetic field is not parallel to the spin magnetization and a local spin torque is present in the ground state of the Kohn-Sham system. As a proof of principle, we apply the functional to a Chromium monolayer in the noncollinear 120°-Néel state.

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Since the discovery of giant magnetoresistance [1], the field of spintronics [2] has played an important role in the everlasting goal to miniaturize devices for data storage and manipulation. For instance, the coupling of orbital and spin degrees of freedom is used to move magnetic domain walls in so-called “racetrack” memory devices [3] via a charge current. Similarly, spin polarized currents can switch the magnetic state of spin valves by means of the so-called spin-transfer torque [4]. Whenever spin-orbit coupling is present, there is no global spin-quantization axis and the spin magnetization becomes noncollinear. A specific example of noncollinear magnetic structures on the nanoscale are skyrmions [5], i.e., topological twists in the spin magnetization, which recently have been observed in magnetic solids [6] and magnetic surfaces [7]. Even within a single atom, noncollinear magnetism is present [8].

Density-functional theory (DFT) [9] is presently the most widely used approach for determining the electronic structure of large molecules and solids. Shortly after the original formulation by Hohenberg and Kohn [10] in terms of the electronic density $n(\mathbf{r})$ alone, the theory was extended to include also the spin magnetization $\mathbf{m}(\mathbf{r})$ as a fundamental variable [11]. Spin-density-functional theory (SDFT) applies to Hamiltonians of the form

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

where in addition to the kinetic energy $\hat{\mathcal{T}}$, the potential energy $\hat{\mathcal{V}}$ and the electron-electron interaction energy $\hat{\mathcal{W}}$ a contribution, $\hat{\mathcal{B}}$, due to an external magnetic field $\mathbf{B}(\mathbf{r})$ is considered, i.e.,

$$\hat{\mathcal{B}} = -\mu_B \int d^3r \mathbf{B}(\mathbf{r}) \cdot \hat{\mathbf{m}}(\mathbf{r}). \quad (2)$$

The operator $\hat{\mathbf{m}}(\mathbf{r}) = \hat{\Phi}^\dagger(\mathbf{r}) \boldsymbol{\sigma} \hat{\Phi}(\mathbf{r})$, representing the spin magnetization, is defined in terms of the spinor field

$\hat{\Phi}^\dagger(\mathbf{r}) = (\hat{\phi}_\uparrow^\dagger(\mathbf{r}) \hat{\phi}_\downarrow^\dagger(\mathbf{r}))$ and the vector of Pauli matrices $\boldsymbol{\sigma}$.

An immediate application of SDFT is to find the configuration of the spin magnetization which is lowest in energy and, hence, the most stable. Furthermore, it is possible to shape the spin magnetization via an external magnetic field. Comparing energies for different magnetic configurations (or equivalently different external magnetic fields) one can map out the energy landscape for a given material. A specific example is to constrain the spin magnetization to rotate in space with a given wave vector \mathbf{q} in order to compute the magnon dispersion by $\epsilon_{\text{mag}}(\mathbf{q}) = E(\mathbf{q}) - E(0)$ (frozen magnon approach).

As always in DFTs the success of the theory hinges on the availability of accurate and physically sound approximations to the exchange-correlation (xc) energy $E_{\text{xc}}[n, \mathbf{m}]$ —a functional of $n(\mathbf{r})$ and $\mathbf{m}(\mathbf{r})$ in the case of SDFT. The functional derivative of $E_{\text{xc}}[n, \mathbf{m}]$ with respect to the density (spin magnetization) yields the so-called xc potential $v_{\text{xc}}(\mathbf{r})$ [xc magnetic field $\mathbf{B}_{\text{xc}}(\mathbf{r})$]. These xc potentials describe the effect of exchange and correlation in the Kohn-Sham (KS) system [12], an effective system of noninteracting electrons, exposed to the potential $v_s(\mathbf{r}) = v(\mathbf{r}) + v_H(\mathbf{r}) + v_{\text{xc}}(\mathbf{r})$ and magnetic field $\mathbf{B}_s(\mathbf{r}) = \mathbf{B}(\mathbf{r}) + \mathbf{B}_{\text{xc}}(\mathbf{r})$, which reproduces the density and spin magnetization of the interacting system.

The simplest approximation in the framework of DFT is the local density approximation (LDA) [12], which determines the xc energy of the nonuniform systems by treating it locally as a uniform electron gas. Including the spin magnetization, this idea is readily generalized yielding for the xc energy

$$E_{\text{xc}}^{\text{LSDA}}[n, \mathbf{m}] = \int d^3r n(\mathbf{r}) \epsilon_{\text{xc}}^{\text{unif}}(n(\mathbf{r}), m(\mathbf{r})), \quad (3)$$

the so-called local spin density approximation (LSDA), with $m(\mathbf{r})$ being the magnitude of $\mathbf{m}(\mathbf{r})$ and $\epsilon_{\text{xc}}^{\text{unif}}$ the xc

energy of a spin-polarized uniform electron gas (UEG). For collinear magnetism, i.e., $\mathbf{m}(\mathbf{r})$ pointing in the same direction everywhere in space, a plethora of functionals was derived (cf. Ref. [13]) improving over the collinear LSDA; however, much less is known about constructing functionals for noncollinear magnetism, where the direction of $\mathbf{m}(\mathbf{r})$ is allowed to vary freely in space. In fact, most applications of noncollinear SDFT to date are based on the idea, pioneered by Kübler *et al.* [14], to apply collinear functionals to noncollinear systems by evaluating the functional in a local reference frame with the local z axis determined by the direction of $\mathbf{m}(\mathbf{r})$. The LSDA, defined in Eq. (3), employs a local reference frame intrinsically which can be seen by evaluating the corresponding xc magnetic field

$$\mathbf{B}_{\text{xc}}^{\text{LSDA}}(\mathbf{r}) = -\frac{\delta E_{\text{xc}}^{\text{LSDA}}[n, \mathbf{m}]}{\delta \mu_{\text{B}} \mathbf{m}(\mathbf{r})} = -\frac{\partial \varepsilon_{\text{xc}}^{\text{unif}}}{\partial m} \frac{n(\mathbf{r}) \mathbf{m}(\mathbf{r})}{\mu_{\text{B}} m(\mathbf{r})}. \quad (4)$$

By construction, $\mathbf{B}_{\text{xc}}^{\text{LSDA}}$ is always aligned with \mathbf{m} . The same is true for generalized gradient approximations (GGAs) employing the aforementioned rotation to a local reference frame. In recent years, attempts were made to extend GGAs and meta-GGAs to noncollinear systems without invoking a local reference frame in order to produce a $\mathbf{B}_{\text{xc}}(\mathbf{r})$ which is noncollinear with respect to $\mathbf{m}(\mathbf{r})$ [15]. Since collinear functionals are usually formulated in terms of $n_{\uparrow}(\mathbf{r})$ and $n_{\downarrow}(\mathbf{r})$ [as opposed to $n(\mathbf{r})$ and $\mathbf{m}(\mathbf{r})$] and gradients thereof, these approaches require a prescription mapping the gradient of $\mathbf{m}(\mathbf{r})$ (a 3×3 -matrix for noncollinear systems) to gradients of $n_{\uparrow}(\mathbf{r})$ and $n_{\downarrow}(\mathbf{r})$. Sharma *et al.* demonstrated that orbital functionals yield, in general, a \mathbf{B}_{xc} which is noncollinear with respect to \mathbf{m} [16]. Another approach was to consider the variations of the direction of $\mathbf{m}(\mathbf{r})$ perturbatively [17,18]. Capelle and Oliveira proposed a nonlocal DFT approach [19,20], in close analogy to the DFT for superconductors [21].

In this Letter, we show that the very idea of the LSDA can be extended in a nonperturbative way to yield a new functional for SDFT depending on transverse gradients. This means that the xc functional depends on spatial variations of the direction of \mathbf{m} and, as a consequence, the xc magnetic field exerts a local torque on the spin magnetization. This local torque is important for the *ab initio* description of spin dynamics [22].

In the LSDA, the spin polarized UEG is chosen as a reference system to determine the local xc energy. Note that the LSDA does not employ the ground-state xc energy of the UEG, but instead the minimal xc energy of the UEG under the constraint that its spin magnetization is m_0 . Usually, one imposes the constraint of a fixed spin magnetization via a uniform magnetic field. The new functional is based on the idea of considering a reference system with a noncollinear spin magnetization. In close analogy to the LSDA, the local xc energy is determined from the UEG constrained to be in the so-called spin-spiral-wave (SSW)

state [23]. The SSW state of the UEG is characterized by a constant density n_0 and a spin magnetization of the form

$$\mathbf{m}_0(\mathbf{r}) = m_0 \begin{pmatrix} s \cos(\mathbf{q} \cdot \mathbf{r}) \\ s \sin(\mathbf{q} \cdot \mathbf{r}) \\ \sqrt{1-s^2} \end{pmatrix}, \quad (5)$$

with $s = \sin(\theta)$ and θ is the azimuthal angle between the rotating part (in the x - y plane) and the constant part (parallel to the z axis). Similar to the case of the uniformly polarized UEG, the constraint of a spin-spiral magnetization is imposed via a local external magnetic field that itself has a spiral structure [24]. The xc energy of the SSW UEG depends on four parameters: n_0 , m_0 , s , and $q = |\mathbf{q}|$. As we will see below, it is possible to define local $s(\mathbf{r})$ and $q(\mathbf{r})$ in terms of transverse gradients of $\mathbf{m}(\mathbf{r})$ which leads to the definition of the SSW functional

$$E_{\text{xc}}^{\text{SSW}}[n, \mathbf{m}] = \int d^3r n(\mathbf{r}) \varepsilon_{\text{xc}}^{\text{SSW}}(n(\mathbf{r}), m(\mathbf{r}), s(\mathbf{r}), q(\mathbf{r})), \quad (6)$$

where $\varepsilon_{\text{xc}}^{\text{SSW}}(n, m, s, q)$ is the minimal xc energy of the UEG under the constraint that it is in the SSW state specified by n , m , s , and q . It is important to realize that the LSDA is included in this definition in the limits $s \rightarrow 0$ or $q \rightarrow 0$, i.e., $\varepsilon_{\text{xc}}^{\text{SSW}}(n, m, s, q = 0) = \varepsilon_{\text{xc}}^{\text{SSW}}(n, m, s = 0, q) = \varepsilon_{\text{xc}}^{\text{SSW}}(n, m, s = 0, q = 0) = \varepsilon_{\text{xc}}^{\text{unif}}(n, m)$. This can be emphasized by rewriting the SSW functional as

$$E_{\text{xc}}^{\text{SSW}}[n, \mathbf{m}] = \int d^3r n(\mathbf{r}) \varepsilon_{\text{xc}}^{\text{unif}}(n(\mathbf{r}), m(\mathbf{r})) \times \{1 + S_{\text{xc}}(n(\mathbf{r}), m(\mathbf{r}), s(\mathbf{r}), q(\mathbf{r}))\}, \quad (7)$$

where we have introduced the spin gradient enhancement (SGE)

$$S_{\text{xc}}(n, m, s, q) = \frac{\varepsilon_{\text{xc}}^{\text{SSW}}(n, m, s, q) - \varepsilon_{\text{xc}}^{\text{unif}}(n, m)}{\varepsilon_{\text{xc}}^{\text{unif}}(n, m)}. \quad (8)$$

Before we discuss the explicit form of the local $s(\mathbf{r})$ and $q(\mathbf{r})$, we briefly discuss global, i.e., spatially independent, rotations of the internal (spin) space. These rotations correspond to transforming $\hat{\Phi}(\mathbf{r}) \rightarrow \mathcal{U} \hat{\Phi}(\mathbf{r})$, where \mathcal{U} is an element of SU(2) (a rotation of the internal [spin] degree of freedom). Note that spatial vectors, e.g., the (charge) current $\mathbf{j}(\mathbf{r})$, are invariant under such internal rotations whereas spin vectors as $\mathbf{m}(\mathbf{r})$ transform as $\mathbf{m}(\mathbf{r}) \rightarrow \underline{R} \mathbf{m}(\mathbf{r})$, with \underline{R} being the 3×3 rotation matrix corresponding to \mathcal{U} . Since the kinetic energy \hat{T} and the interaction energy \hat{W} are invariant under global rotations of the internal space, it follows that $E_{\text{xc}}[n, \mathbf{m}] = E_{\text{xc}}[n, \underline{R} \mathbf{m}]$. Considering infinitesimal spin rotations, one obtains the so-called zero-torque theorem

$$0 = \int d^3r \mathbf{m}(\mathbf{r}) \times \mathbf{B}_{\text{xc}}(\mathbf{r}), \quad (9)$$

which was first derived by Capelle *et al.* via the equation of motion for the spin magnetization [22]. It states that \mathbf{B}_{xc} cannot exert a net torque on the whole system.

A simple rule to follow in order to ensure that explicit functionals for SDFT obey the zero-torque theorem is to write $E_{xc}[n, \mathbf{m}]$ in terms of proper scalars; i.e., spin indices have to be contracted with spin indices and spatial indices with spatial indices. This implies that the determination of the local xc energy in terms of strictly local densities is exhausted by $n(\mathbf{r})$ and $m(\mathbf{r})$. Hence, the local $s(\mathbf{r})$ and $q(\mathbf{r})$ have to be determined from properly contracted gradients of $\mathbf{m}(\mathbf{r})$.

Let us first look at $D(\mathbf{r}) = |\nabla \otimes \mathbf{m}(\mathbf{r})|^2$, which corresponds to the total first order change of $\mathbf{m}(\mathbf{r})$. It can be split into a longitudinal contribution $D_L(\mathbf{r})$ and a transverse contribution $D_T(\mathbf{r})$, i.e.,

$$D(\mathbf{r}) = \frac{1}{m^2(\mathbf{r})}(D_L(\mathbf{r}) + D_T(\mathbf{r})), \quad (10)$$

$$D_L(\mathbf{r}) = |\mathbf{m}(\mathbf{r}) \cdot (\nabla \otimes \mathbf{m}(\mathbf{r}))|^2, \quad (11)$$

$$D_T(\mathbf{r}) = |\mathbf{m}(\mathbf{r}) \times (\nabla \otimes \mathbf{m}(\mathbf{r}))|^2, \quad (12)$$

where the meaning of longitudinal and transverse is defined by the local direction of $\mathbf{m}(\mathbf{r})$. We use “ \otimes ” to emphasize that the gradient of the magnetization is a tensor. The scalar and the cross product in Eqs. (11) and (12) act on the components of \mathbf{m} [25]. For the SSW UEG, the two contributions are $D_L^{SSW} = 0$ and $D_T^{SSW} = m_0^4 s^2 q^2$. Both contributions are constant in space for the SSW UEG and hence play a similar role as the density n_0 and the magnitude of the spin magnetization m_0 ; i.e., they locally characterize the state. D_L^{SSW} vanishes because the spin magnetization in the SSW UEG only rotates (the magnitude m is constant). D_T does not vanish, but it only determines the combination sq .

Accordingly, we look at the second order variation $d(\mathbf{r}) = |\nabla^2 \mathbf{m}(\mathbf{r})|^2$. Again, it can be analyzed with respect to longitudinal and transverse contributions

$$d(\mathbf{r}) = \frac{1}{m^2(\mathbf{r})}(d_L(\mathbf{r}) + d_T(\mathbf{r})), \quad (13)$$

$$d_L(\mathbf{r}) = |\mathbf{m}(\mathbf{r}) \cdot (\nabla^2 \mathbf{m}(\mathbf{r}))|^2, \quad (14)$$

$$d_T(\mathbf{r}) = |\mathbf{m}(\mathbf{r}) \times (\nabla^2 \mathbf{m}(\mathbf{r}))|^2. \quad (15)$$

For our reference system, this yields $d_L^{SSW} = m_0^4 s^4 q^4$ and $d_T^{SSW} = m_0^4 (1 - s^2) s^2 q^4$. The change of $\mathbf{m}(\mathbf{r})$ to first order is perpendicular to $\mathbf{m}(\mathbf{r})$, but to second order $\mathbf{m}(\mathbf{r})$ also changes in the direction of $\mathbf{m}(\mathbf{r})$ which explains why d_L^{SSW} does not vanish for the SSW UEG. However, we see that d_L^{SSW} provides the same information as D_T^{SSW} , meaning, sq to some power. Adopting the strategy that we obtain the characteristic parameters for the local xc energy choosing

the order of derivatives as low as possible, $s(\mathbf{r})$ and $q(\mathbf{r})$ are given by

$$s(\mathbf{r}) = \sqrt{\frac{D_T^2(\mathbf{r})}{D_T^2(\mathbf{r}) + m^4(\mathbf{r})d_T(\mathbf{r})}}, \quad (16)$$

$$q(\mathbf{r}) = \sqrt{\frac{D_T^2(\mathbf{r}) + m^4(\mathbf{r})d_T(\mathbf{r})}{m^4(\mathbf{r})D_T(\mathbf{r})}}. \quad (17)$$

This completes the definition of the SSW functional Eq. (6), or equivalently, the SGE to the LSDA Eq. (7).

By definition [c.f. Eq. (16)] the local $s(\mathbf{r})$ is between [0, 1] in accordance with being the sine of an azimuthal angle. Furthermore, we have the following hierarchy in the dependence of the SGE, Eq. (7), on the transverse gradients: (i) If $D_T(\mathbf{r}) = 0$, the SGE correction is zero. (ii) If $D_T(\mathbf{r}) \neq 0$ and $d_T(\mathbf{r}) = 0$, the SGE correction is obtained from a planar SSW ($s = 1$). (iii) If both transverse gradients are nonzero, the SGE correction is obtained from a general SSW.

We proceed by evaluating the xc magnetic field from the SSW functional,

$$\mathbf{B}_{xc}^{SSW}(\mathbf{r}) = -\frac{\delta E_{xc}[n, \mathbf{m}]}{\delta \mu_B \mathbf{m}(\mathbf{r})} = \mathbf{B}_{xc}^m + \mathbf{B}_{xc}^{D_T} + \mathbf{B}_{xc}^{d_T}, \quad (18)$$

where we split $\mathbf{B}_{xc}^{SSW}(\mathbf{r})$ into contributions coming from the dependence of ε_{xc}^{SSW} on m , D_T , and d_T , respectively. The explicit evaluation of $\mathbf{B}_{xc}^{SSW}(\mathbf{r})$ is straightforward, but rather lengthy. Here, we will show the energetic content in the KS system, i.e.,

$$E_{B_{xc}}^{KS} = -\mu_B \int d^3r m(\mathbf{r}) \cdot \mathbf{B}_{xc}(\mathbf{r}) \\ = 2 \int d^3r n(\mathbf{r}) (\partial_{m^2} \varepsilon_{xc}^{SSW}) |\mathbf{m}(\mathbf{r})|^2 \quad (19)$$

$$+ 4 \int d^3r n(\mathbf{r}) (\partial_{D_T} \varepsilon_{xc}^{SSW}) |\mathbf{m}(\mathbf{r}) \times (\nabla \otimes \mathbf{m}(\mathbf{r}))|^2 \quad (20)$$

$$+ 4 \int d^3r n(\mathbf{r}) (\partial_{d_T} \varepsilon_{xc}^{SSW}) |\mathbf{m}(\mathbf{r}) \times (\nabla^2 \mathbf{m}(\mathbf{r}))|^2. \quad (21)$$

The first term [Eq. (19)] is already present in the LSDA, whereas the other two terms [Eqs. (20) and (21)] arise due to the inclusion of the SGE. The zero-torque theorem, Eq. (9), is fulfilled by construction; however, the new terms in $\mathbf{B}_{xc}(\mathbf{r})$ are noncollinear with respect to $\mathbf{m}(\mathbf{r})$; i.e., they provide a local torque.

The final step for a practical implementation of the SSW functional is the determination of the SGE from the SSW UEG. We have evaluated $S_{xc}(n, m, s, q)$ using the random-phase approximation (RPA) for the SSW UEG. It is important to stress that we approximate the S_{xc} with the RPA and not ε_{xc}^{SSW} . In this way, the SSW functional reduces to the LSDA parametrized using the Monte Carlo reference data

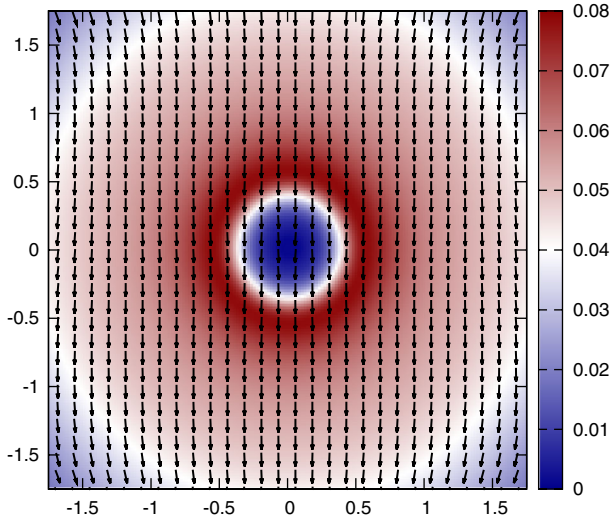


FIG. 1 (color online). Magnitude (color coded) and direction (arrows) of $\mathbf{B}_{xc}(\mathbf{r})$ around an atom for the Cr monolayer using the LSDA.

[26]. From ~ 65000 data points in the four-dimensional domain of S_{xc} we have constructed a polynomial fit for S_{xc} .

As a first application, we have implemented the SSW functional in the ELK code [27] in order to investigate the chromium monolayer in the 120° -Néel state. In Figs. 1 and 2, we plot the magnitude and direction of \mathbf{B}_{xc} in order to illustrate the qualitative difference between the LSDA and the SSW functional. While the local spin magnetizations $\mathbf{m}(\mathbf{r})$ are similar for the LSDA and the SSW functional, \mathbf{B}_{xc} obtained via the SGE exhibits much more structure compared to the LSDA \mathbf{B}_{xc} . As a result, the local torque does not vanish and a ground-state spin current is present in the KS system [28]. The local torque which is

completely missed by the usual LSDA is shown explicitly in Fig. 3 for the noncollinear 120° -Néel state. The global zero-torque theorem [cf. Eq. (9)] may be inferred from the pattern of negative (blue) and positive (red) local torques around the nuclei. Since the SSW functional is not restricted to small $q(\mathbf{r})$, it accounts for the intra-atomic noncollinearity.

In conclusion, we have proposed a novel functional for SDFT depending on the first and second order transverse gradients of $\mathbf{m}(\mathbf{r})$. We emphasize that this functional is formulated in terms of an enhancement to the LSDA. In particular, this means that the correction vanishes in the case of a collinear system. The construction of the new functional parallels closely the original formulation of the LSDA. On one hand, this means that the system is locally treated as a uniform electron gas in the SSW state, which may appear as a rather crude approximation. On the other hand, the success of DFT may be attributed, to some extent, to the fact that the LDA already represents a reasonable approximation even for strongly inhomogeneous systems. GGAs are also corrections to the LSDA; hence, it is conceivable to employ the two corrections simultaneously. Since GGAs are constructed having collinear systems in mind, one may argue that the longitudinal gradients $D_L(\mathbf{r})$, $d_L(\mathbf{r})$ should enter in the GGA part. We expect that the SGE will improve the *ab initio* description of materials exhibiting a noncollinear magnetic structure.

While the corrections to the part of the $\mathbf{B}_{xc}(\mathbf{r})$ parallel to $\mathbf{m}(\mathbf{r})$ will adjust the energetics, the perpendicular part of $\mathbf{B}_{xc}(\mathbf{r})$ describes the xc corrections to the spin current, which in turn is crucial for *ab initio* spin dynamics. We expect that the functional presented in this Letter will pave the road to a better description of domain wall motion

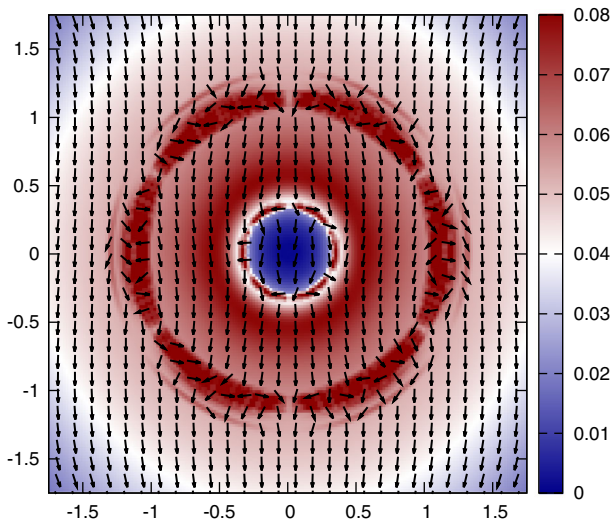


FIG. 2 (color online). Same as Fig. 1 for the SSW functional (LSDA + SGE). Note the richer structure of $\mathbf{B}_{xc}(\mathbf{r})$ close to the Cr nucleus.

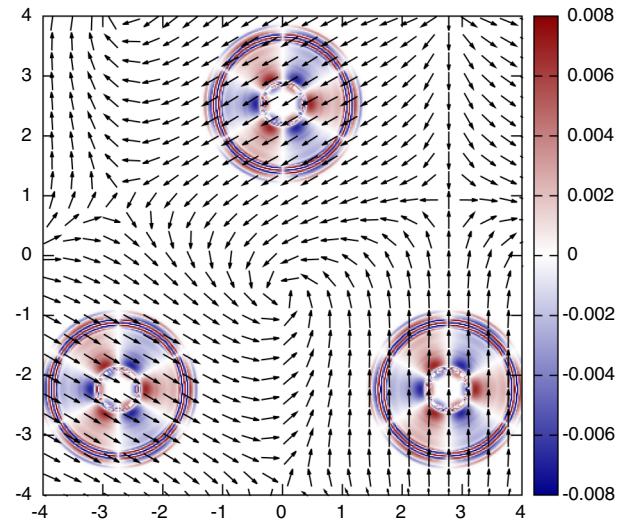


FIG. 3 (color online). The z component of $\mathbf{m}(\mathbf{r}) \times \mathbf{B}_{xc}(\mathbf{r})$ (color coded) around Cr atoms in the monolayer computed using the SGE functional. The arrows show the direction of the spin magnetization.

and spin wave propagation from first principles in the framework of time-dependent SDFT.

In both aforementioned scenarios, it is important to have a numerically accessible functional which, given currently available computing facilities, implies the use of semilocal functionals. We have demonstrated that noncollinearity can be included by a generalization of the reference system employed in the LSDA, and hence, the numerical accessibility of the LSDA is retained in the SSW functional making it the ideal candidate for large scale quantum simulations.

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- [24] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevLett.111.156401> where this is shown explicitly for a noninteracting electron gas.
- [25] Note that “ $|\dots|^2$ ” always implies a contraction of the remaining indices, e.g., $|\nabla \otimes \mathbf{m}|^2 = (\partial_i m_j)(\partial_i m_j)$ where a summation of repeated indices is implied.
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- [28] This can be seen from the equation of motion for the spin magnetization (cf. Ref. [22]).