Exchange torque in noncollinear spin density functional theory with a semilocal exchange functional

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We present a new semilocal exchange energy functional for spin density functional theory (SDFT) based on a short-range expansion of the spin-resolved exchange hole. Our exchange functional is directly derived for noncollinear magnetism, U(1) and SU(2) gauge invariant, and gives rise to non-vanishing exchange torques. The functional is tested for frustrated antiferromagnetic chromium clusters and shown to perform favorably compared to the far more expensive Slater potential and optimized effective potential for exact exchange. This provides a path forward for functional development in noncollinear SDFT and the *ab initio* study of magnetic materials in and out of equilibrium.

In the framework of density functional theories (DFTs), the key to success in describing the equilibrium and time-dependent behavior of an electronic system, such as its electronic or magnetic properties, is inherently related to the quality of approximations of the exchange-correlation (xc) energy and its functional derivatives. In the quest to develop more accurate xc functionals, most of the recent effort has been devoted to the construction of approximations for spin unpolarized systems or for systems well described by collinear spins [1–3].

Considering the ever growing interest in the fields of spintronics and optical control of magnetism, reliable functionals going beyond going beyond the widely used local spin density approximation (LSDA)are highly desirable. In particular, one of the features expected from the exact xc magnetic field ($\mathbf{B}_{\mathrm{xc}}$) of spin density functional theory (SDFT) [4, 5] is to generate locally a non-zero xc torque, which arises from the fact that in SDFT the Kohn-Sham current is not the same as the exact many-body current [6]. Globally, this xc torque must average to zero, which is known as the zero-torque theorem [6]. However, the practical significance of xc torques is still relatively little explored [7, 8], and a better understanding would be beneficial for the application of SDFT to noncollinear magnetism and spin dynamics [9].

In frustrated antiferromagnetic systems, such as free-standing Cr monolayers, local xc torques can be sizable in the vicinity of the Cr atoms [10], based on calculations performed at the level of the optimized effective potential (OEP) for exact exchange (EXX) [11, 12]. How this nonzero xc torque affects the static and dynamical properties of magnetic systems, such as light-induced demagnetization [9] or the dispersion of spin waves [13], is an open question that needs to be addressed in order to gain understanding and control of the magnetization dynamics in these systems. More recently, it was demonstrated using a source-free version of the LSDA [14] that the xc torque affects the light-demagnetization process [15].

Two main approaches are available for constructing xc functionals for noncollinear systems. A first approach, which is most commonly employed, consists in promoting existing functionals for collinear systems to work for non-collinear ones. This was initiated by the work of Kübler and others [16, 17], who showed how to carry out non-collinear LSDA calculations by performing a rotation of the spin density matrix in the frame of the local magnetization $\mathbf{m}$. The drawback of this approach is clear: because the resulting local $\mathbf{B}_{\mathrm{xc}}$ is aligned with $\mathbf{m}$, this cannot produce any local xc torques. Various extensions of this approach to generalized spin gradient approximation (GGA) functionals have been proposed since then [18, 19]. While these different schemes can produce nonzero xc torques, they tend to suffer from numerical instabilities and can encounter difficulties in reproducing the collinear limit unless special care is taken [20].

In this work, we follow a second approach for constructing xc functionals for noncollinear magnetism, which consists in developing directly xc functionals for noncollinear systems instead of promoting existing collinear functionals to the noncollinear case. This general philosophy was used in some earlier studies, such as the transverse spin-gradient functional developed by Eich and Gross [21]. More recently, it was proposed to build a meta-GGA (MGGA) functional for noncollinear systems [22] based on the short-range expansion of the exchange hole in the noncollinear case, but no numerical implementation of this was reported so far. Finally, a class of orbital-dependent functionals for noncollinear SDFT was proposed in Ref. 23 and applied to the asymmetric Hubbard dimer and other small lattice systems [8].

In this Letter, we show how to use a short-range expansion of the exchange hole to build an MGGA functional for noncollinear SDFT. We propose an effective noncollinear exchange hole that can be properly approximated by existing models for the exchange hole. From this, we obtain an MGGA exchange functional that not

only depends locally on the direction of $\mathbf{m}$ but also on the direction of the curvature of the exchange hole, which implies that the resulting $\mathbf{B}_{\mathrm{xc}}$ can exert a local torque on $\mathbf{m}$. We then apply this functional to planar Cr clusters with frustrated antiferromagnetic magnetic interactions and show that not only the functional produces a nonzero exchange torque, but that it properly reproduces the most salient features of the exchange torque obtained by the far more complicated noncollinear Slater potential and EXX-OEP. We mention that it was shown for model systems that Slater and EXX-OEP yield decent approximations to the exact xc torques, as long as the systems are not too strongly interacting [8].

Our starting point is the exchange energy of a system of N electrons,

$$E_{\mathrm{x}} = -\frac{1}{2} \int \int \frac{d\mathbf{r}\, d\mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|} \mathrm{Tr}\left[\underline{\underline{\gamma}}(\mathbf{r}, \mathbf{r}')\underline{\underline{\gamma}}(\mathbf{r}', \mathbf{r})\right], \qquad (1)$$

where Tr is the trace over spin indices of the one-particle spin density matrix $\gamma_{\sigma\tau}(\mathbf{r}, \mathbf{r}') = \sum_j^N \psi_{j\sigma}(\mathbf{r})\psi_{j\tau}^*(\mathbf{r}')$, constructed from two-component spinor Kohn-Sham orbitals, where $\sigma = \uparrow, \downarrow$ and likewise for τ. Here and in the following, doubly underlined quantities such as $\underline{\underline{\gamma}}$ represent 2×2 matrices. Here, E_{x} is U(1)$\times$ SU(2) gauge invariant [22]; directly approximating E_{x} is therefore a good strategy to produce meaningful exchange functionals for noncollinear SDFT.

From E_{x} of Eq. (1) it is possible to derive the noncollinear OEP EXX potential, as well as the Slater potential for noncollinear spins [23]. This allows us to generalize the definition of the exchange hole to systems with noncollinear spin, as discussed in the Supplementary Material (SM) [25]. However, the exchange hole is now a 2×2 matrix in spin space, it would be necessary to approximate its diagonal as well as off-diagonal terms. The existing models for the collinear exchange hole [26] rely on the fact that the modeled exchange is positive and normalized to unity. These properties are obviously not fulfilled by the off-diagonal terms of the noncollinear exchange hole, which makes these terms complicated to approximate using existing collinear models.

To circumvent this problem, we seek an alternative quantity to approximate, for which we could use already existing collinear models. For this, we rewrite Eq. (1) as

$$E_{\mathrm{x}} = -\frac{1}{2} \int d\mathbf{r}\, n(\mathbf{r}) \int d\mathbf{r}' \frac{h_{\mathrm{x}}(\mathbf{r}, \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}, \qquad (2)$$

defining $h_{\mathrm{x}}(\mathbf{r}, \mathbf{r}') = \mathrm{Tr}\left[\underline{\underline{\gamma}}(\mathbf{r}, \mathbf{r}')\underline{\underline{\gamma}}(\mathbf{r}', \mathbf{r})\right]/n(\mathbf{r})$, where $n(\mathbf{r}) = \sum_\sigma n_{\sigma\sigma}(\mathbf{r})$ is the total density. The quantity h_{x}, which we refer in the following as the effective exchange hole, displays the same properties as the physical exchange hole for unpolarized or collinear-spin systems. Indeed, $\mathrm{Tr}\left[\underline{\underline{\gamma}}(\mathbf{r}, \mathbf{r}')\underline{\underline{\gamma}}(\mathbf{r}', \mathbf{r})\right]$ is always positive, and hence h_{x} is always positive, too. Moreover, h_{x} has the right normalization condition, i.e., $\int d\mathbf{r}' h_{\mathrm{x}}(\mathbf{r}, \mathbf{r}') = 1$. Therefore, h_{x} is suited to be approximated by already existing

models for the collinear exchange hole. A recent review [26] of the existing models for the collinear exchange hole found that the hydrogenic model [27] seems to perform best, at least for the systems considered. We therefore employ this model in the following to approximate h_{x} and construct a noncollinear MGGA exchange functional.

Only the spherical average of the effective exchange-hole function around a given reference point $\mathbf{r}$ is relevant for the exchange energy. We therefore perform a Taylor expansion of the spherical average of h_{x} to obtain its short-range behavior around a reference point $\mathbf{r}$, which gives, up to second order in the distance s from $\mathbf{r}$, its on-top value and its curvature, see SM [25]. Omitting the explicit dependence on $\mathbf{r}$, we obtain, up to second order in s,

$$h_{\mathrm{x}}(s) = \sum_\sigma \frac{n_{\sigma\sigma}^2 + |n_{\sigma-\sigma}|^2}{n} + \frac{s^2}{6n} \sum_\sigma \Big[n_{\sigma\sigma} \nabla^2 n_{\sigma\sigma}$$
$$+ \Re(n_{\sigma-\sigma}\nabla^2 n_{-\sigma\sigma}) - 2n_{\sigma\sigma}\tilde{\tau}_{\sigma\sigma} - 2\Re(n_{\sigma-\sigma}\tilde{\tau}_{-\sigma\sigma})$$
$$+ \frac{1}{2}(|\nabla n_{\sigma\sigma}|^2 + |\nabla n_{\sigma-\sigma}|^2) \Big] + \mathcal{O}(s^4). \qquad (3)$$

Here, $\tilde{\tau}$ is an effective kinetic energy density defined by the matrix equation

$$\underline{\underline{n}}\,\underline{\underline{\tilde{\tau}}} + \underline{\underline{\tilde{\tau}}}\,\underline{\underline{n}} = \underline{\underline{n}}\,\underline{\underline{\tau}} + \underline{\underline{\tau}}\,\underline{\underline{n}} - 2i\nabla(\underline{\underline{n}}\,\underline{\underline{j}} - \underline{\underline{j}}\,\underline{\underline{n}}) - 2\underline{\underline{\mathbf{j}}} \cdot \underline{\underline{\mathbf{j}}}, \qquad (4)$$

with $\tau_{\alpha\beta} = \sum_j (\nabla\psi_{j\alpha}(\mathbf{r})) \cdot \nabla\psi_{j\beta}^*(\mathbf{r})$ the usual kinetic energy density, and $\underline{\underline{\mathbf{j}}}$ is the paramagnetic current density. The quantity $\tilde{\tau}$ is U(1) gauge invariant but is not invariant under a local SU(2) gauge transformation [22]. Of course, the entire curvature is SU(2) invariant. Thus, in order to obtain an expression for the spherically averaged effective exchange hole that is made of SU(2) invariant building blocks, we rewrite $h_{\mathrm{x}}(s)$ as

$$h_{\mathrm{x}}(s) = \left(\frac{n}{2} + \frac{|\mathbf{m}|^2}{2n}\right) + \frac{s^2}{6}\left[\nabla^2 n - 2\gamma\left(\bar{\tau} - \tau^W\right)\right]$$
$$= \left(\frac{n}{2} + \frac{|\mathbf{m}|^2}{2n}\right)\Big[1 + s^2 k_{\mathrm{F}}^2$$
$$\times \left(\frac{2}{3}q(n, |\mathbf{m}|) - \frac{\gamma}{5}\alpha(n, |\mathbf{m}|, |\nabla n|, \bar{\tau})\right)\Big], \qquad (5)$$

where $\bar{\tau}$ is given by

$$2n\bar{\tau} = \mathrm{Tr}\left[\underline{\underline{n}}\,\underline{\underline{\tilde{\tau}}} + \underline{\underline{\tilde{\tau}}}\,\underline{\underline{n}}\right] + \sum_\sigma \Big[n_{\sigma\sigma}\nabla^2 n_{-\sigma-\sigma} \qquad (6)$$
$$- \Re(n_{\sigma-\sigma}\nabla^2 n_{-\sigma\sigma}) - \frac{|\nabla n_{\sigma-\sigma}|^2}{2} + \frac{\nabla n_{\sigma\sigma} \cdot \nabla n_{-\sigma-\sigma}}{2} \Big].$$

One can show (see SM [25]) that in the one-electron limit $\bar{\tau}$ reduces to the von Weizsäcker kinetic-energy density $\tau^W = |\nabla n|^2/4n$ and in the uniform gas limit $\bar{\tau}$ reduces to $\bar{\tau}^{\mathrm{unif}} = \frac{3}{5}k_{\mathrm{F}}^2(n/2 + |\mathbf{m}|^2/2n)$, with $k_{\mathrm{F}} = (6\pi^2 n)^{1/3}$. Similar to common collinear MGGAs [28, 29], we introduce a dimensionless parameter $\alpha(n, |\mathbf{m}|, |\nabla n|, \bar{\tau}) = (\bar{\tau} - \tau^W)/\bar{\tau}^{\mathrm{unif}}$, which in the one-electron and uniform gas limits reduces to $\alpha = 0$ and $\alpha = 1$, respectively, as in

the collinear case. We also define a dimensionless Laplacian $q(n, |\mathbf{m}|) = \frac{\nabla^2 n}{4k_F^2(n/2+|\mathbf{m}|^2/2n)}$. The different components of the exchange-hole curvature, in particular $\bar{\tau}$, are all U(1) and SU(2) gauge invariant. Note that the SU(2) gauge invariance implies $\alpha \geq 0$.

Let us comment here on the importance of the gauge invariance. Already at the level of collinear MGGAs, the U(1) gauge invariance is very important, especially for the dynamical case in which the current is non-zero or for the description of current-carrying states, as realized by several authors [30–33]. For noncollinear systems, a semi-relativistic theory including on equal footing electromagnetic fields and spin-related terms in the Hamiltonian (Zeeman term, spin-orbit coupling) should also preserve the local SU(2) gauge invariance [22]. This is the case of the exact-exchange energy, but also in our proposed functional.

It is clear that building a functional from the quantities $(\alpha, q, \bar{\tau})$ will directly recover the collinear limit, which is a strong requirement for any noncollinear xc functional. Moreover, we see that while the on-top term is determined by the direction of $\mathbf{m}$, the curvature has its own direction, which is independent of that of $\mathbf{m}$. This implies that the resulting $\mathbf{B}_{xc}$ is in general not aligned with $\mathbf{m}$, thus producing a nonvanishing local exchange torque. Finally, note that we introduced a scaling factor γ in Eq. (5) to ensure the correct homogeneous electron gas limit, as done in the collinear case [27]. This scaling factor does not break the gauge invariance of the energy, as it acts on a building block which is gauge invariant by itself. The limit of hydrogenic systems ($\alpha = 0$) is therefore not affected by the choice of γ. In the following, we use $\gamma = 0.8$ unless stated otherwise.

Having at hand the short-range behavior of the effective exchange hole, we now express it via a hydrogenic model [27]:

$$\rho_H^x(\mathbf{r}; s) = \frac{a(\mathbf{r})}{16\pi s}\Big[(a(\mathbf{r})|b(\mathbf{r}) - s| + 1)e^{-a(\mathbf{r})|b(\mathbf{r})-s|} - (a(\mathbf{r})(|b(\mathbf{r}) + s| + 1)e^{-a(\mathbf{r})|b(\mathbf{r})+s|}\Big], \quad (7)$$

where $a(\mathbf{r})$ and $b(\mathbf{r})$ are positive parameters of the model that need to be determined. The equations for $a(\mathbf{r})$ and $b(\mathbf{r})$ are found to be identical to the collinear case, with the difference that the on-top part of the exchange hole is not given by the density (see SM [25] for more details). Once these parameters are determined for each $\mathbf{r}$, we obtain the final expression of our noncollinear exchange energy functional:

$$E_x = -\int d\mathbf{r} n(\mathbf{r}) \frac{3(3\pi^2)^{1/3}}{4\pi}\left(n(\mathbf{r}) + \frac{|\mathbf{m}(\mathbf{r})|^2}{n}\right)^{1/3} F_x(\mathbf{r}). \quad (8)$$

Here,

$$F_x(\mathbf{r}) = \frac{2\pi^{2/3}e^{x(\mathbf{r})/3}}{3^{4/3}x(\mathbf{r})}\left[1 - e^{-x(\mathbf{r})}\left(1 + \frac{x(\mathbf{r})}{2}\right)\right] \quad (9)$$

plays the role of an enhancement factor, where $x(\mathbf{r}) = a(\mathbf{r})b(\mathbf{r})$. Importantly, because the on-top effective exchange hole as well as the exchange hole curvature are U(1) and SU(2) gauge invariant, a and b invariant under a local rotation of the spin, and our functional is then U(1) and SU(2) gauge invariant and also recovers naturally the collinear limit. Moreover, because the functional is based on the exchange hole of a physical system (the H atom), the energy is constrained to reasonable values.

By making the total energy stationary with respect to spinor orbital variations, we obtain a differential operator rather than a local multiplicative Kohn-Sham potential, because of the explicit dependence on the kinetic energy density, see SM [25]. This treatment, first used by Neumann, Nobes, and Handy [34], is usually referred as generalized Kohn-Sham (gKS). We refer to MGGA treated using this approach as MGGA-gKS. Alternatively, one can construct a local multiplicative Kohn-Sham potential using the OEP formalism, which we will refer to as MGGA-OEP. This approach tends to be less used as the solution of the OEP equation can be numerically involved [12]. While these two approaches usually give similar total energies, they can yield different results for other quantities of interest such as the nuclear shielding of small molecules [35] or the bandgap of solids [36], and it is therefore interesting to explore how gKS and OEP can differ for noncollinear systems. In order to perform this analysis, we derived an explicit solution for the Krieger-Li-Iafrate (KLI) approximation [11] toward the exact OEP result for noncollinear spins (see SM [25]). The implementation of this solution and of our MGGA functional was done using the real-space code Octopus [37].

As a critical check of the functional, we consider a planar Cr_3 cluster with frustrated antiferromagnetic interactions, which is typically used to test noncollinear versions of collinear functionals [18, 19, 38]. Calculations were performed using a grid spacing of 0.1 Bohr, employing norm-conserving fully relativistic Hartwigsen-Goedecker-Hutter (HGH) pseudo-potentials [39], including semicore electrons as valence ones and spin-orbit coupling in all the simulations. The distance between the Cr atoms is taken to be 3.7 Bohr.

As an important measure of the performance of the functional, we first consider the local magnetic moment of the Cr atoms, see Table I. Interestingly, the exchange-only LSDA (LSDAx) gives much larger magnetic moments than the full LSDA, showing the importance of correlation effects in this system. However, our focus in the present work is an exchange functional. We note that several models have been proposed which locally rescale the exchange hole in order to include correlation effects [40–43]; this might be explored in subsequent work to construct a noncollinear exchange-correlation MGGA.

The proposed MGGA exchange functional significantly improves the description of the magnetic structure of the Cr_3 cluster compared to the LSDAx; all results are rela-
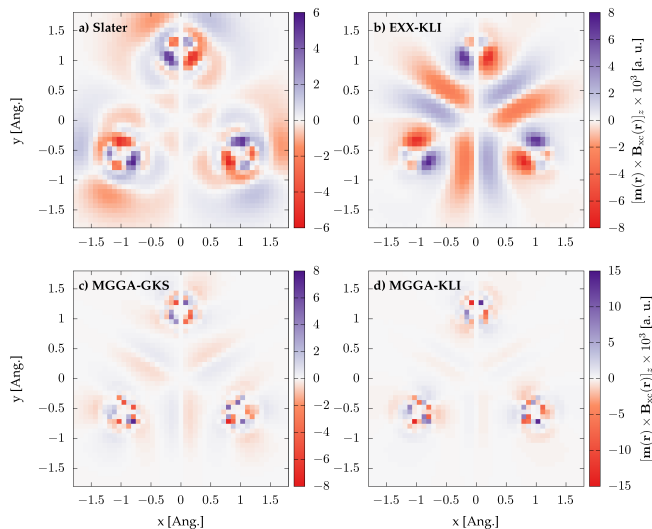
FIG. 1. The z component of the local xc torque $\mathbf{m}(\mathbf{r}) \times \mathbf{B}_{xc}(\mathbf{r})$ around the Cr atoms in a Cr$_3$ cluster, computed from a) Slater potential, b) EXX-KLI, c) MGGA functional at the gKS level, d) MGGA at the OEP-KLI level.

TABLE I. Local magnetic moment m, in μ_B, and ionization potential I_p, in eV, of the Cr atoms in Cr$_3$ obtained for different levels of theory (see text). The moments are obtained by integrating the density in a sphere of radius 1.8 Bohr around the atoms.

Functional	m	I_p
LSDA	1.67	2.90
LSDAx	2.66	2.30
MGGA-gKS ($\gamma = 0.8$)	3.04	3.65
MGGA-gKS ($\gamma = 1$)	3.07	3.53
MGGA-KLI ($\gamma = 0.8$)	3.09	3.59
MGGA-KLI ($\gamma = 1$)	3.14	3.47
Slater	3.48	6.52
EXX-KLI	3.81	4.68
Hartree-Fock	3.86	4.86

tively insensitive to the choice of γ. We note that there is indeed a difference between using the MGGA functional within the gKS framework and using the KLI scheme. This observation is in line with previously reported results for magnetic moments in solids [44, 45], in which it was found that MGGA for collinear magnetism only slightly changes the magnetic moments compared to the ones obtained by GGAs. These two works both used the gKS approach. Our results for Cr$_3$ reveal that more detailed studies of the impact on the choice of gKS versus OEP for treating MGGA in magnetic systems is needed. We also report in Table I the ionization potential computed from the different functionals. Similar conclusions are obtained as from the magnetic moments.

Next, we analyze the exchange torque along the out-of-plane direction of the cluster (the z direction). Figure 1 shows the torque obtained with our MGGA, compared

with Slater and EXX-KLI. Clearly, while the Slater potential provides decent magnetic moments, the exchange torque does not resemble that obtained by EXX-KLI, apart from regions close to the atoms. As expected for our proposed MGGA, we obtain a nonzero exchange torque as a consequence of the fact that the curvature of the noncollinear exchange hole is not aligned with the magnetization direction. The alternation of positive and negative local torques leads to an overall zero torque, as required by the zero-torque theorem [46]. Overall, our functional agrees well with the Slater/EXX-KLI torques in the regions around the atoms. In fact, our MGGA yields fewer torque features than Slater, in better agreement with EXX-KLI, especially in the interstitial region, except that the signs of the features are inverted. There are slight differences between the gKS and OEP implementations of the MGGA, but these are small compared to the differences between MGGA and Slater/EXX-KLI. Further away from the atoms, it is clear that the proposed MGGA does not capture all the details of the exchange torque, as expected for a functional based on a short-range expansion of the noncollinear exchange hole.

It is important to note that compared to the spin-spiral wave noncollinear functional proposed by Eich and Gross [21], which produces a six-fold symmetric exchange torque around all the Cr atoms, our functional yields a torque that depends on the local environment of the atoms, as obtained by the Slater potential, EXX-KLI, or EXX-OEP for Cr monolayers [10]. We can also compare with the noncollinear GGA by Scalmani and Frisch [19], where the exchange torque is also found to depend on the local environment of the atoms, but their result for the torques displays a wrong number of positive-negative features around the atoms compared to those of Slater, EXX-KLI, and our MGGA. Overall, while our MGGA results for Cr$_3$ leave some room for improvement, they do provide a realistic description of the exchange torques.

We also performed calculations for a Cr$_5$ cluster, see Fig. 2, using a distance of 3.7 Bohr between the Cr atoms. Similar to the case of Cr$_3$, our functional produces an exchange torque of the same order of magnitude and with similar features around the atoms as that obtained by Slater or EXX-KLI. As for Cr$_3$, the sign of the exchange torque is inverted in the interstitial region. Again, we attribute this to the short-range nature of our expansion, which has difficulties producing the correct sign of the $\mathbf{B}_x$ away from the atomic centers.

In conclusion, we have developed a new semilocal exchange functional for noncollinear SDFT derived from the short-range behavior of the noncollinear exchange hole. The functional, which belongs to the class of MG-GAs, is numerically well behaved and computationally much cheaper than nonlocal exchange functionals of comparable quality (Slater and KLI) that scale quadratically with the number of state and require the calculation of Coulomb integrals for each pair of occupied Kohn-Sham states. Potential applications of this new exchange func-
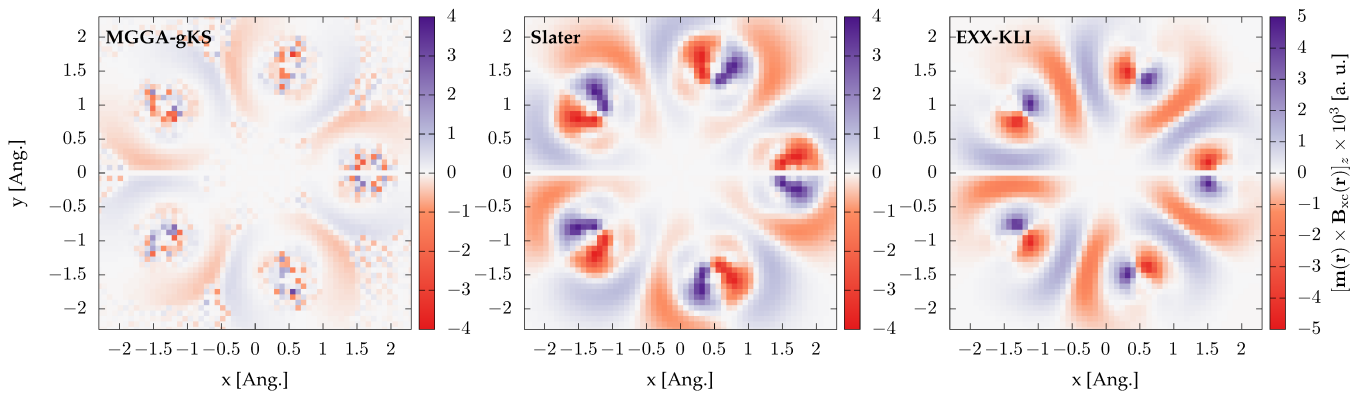
FIG. 2. Out-of-plane component of the exchange torques for a Cr_5 cluster, calculated with MGGA-gKS, Slater, and EXX-KLI.

tional range from *ab initio* equilibrium studies of unconventional spin structures like skyrmions to a variety of dynamical phenomena in magnetic materials such as spin-wave dispersions and the demagnetization in light-driven magnetic solids. From a more fundamental perspective, this work highlights the important and interesting role of xc torques in SDFT, which should motivate further study. Finally we note that while we employed here a specific hydrogenic model for the effective exchange hole, it should be possible to apply our approach to other models and to build new functionals for noncollinear magnetism following similar ideas.

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[25] See Supplemental Material at http://... for the following details: (1) Definition of the noncollinear exchange hole. (2) Hydrogenic model for the exchange hole (3) Derivation of the noncollinear MGGA potential (4) One-electron and uniform gas limit of the exchange hole (5) Explicit solution for the KLI potential. The Supplemental Material includes Ref. [47].

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