First-principles Study of Spin Fluctuations Using Self-adaptive Spin-constrained DFT

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A versatile self-adaptive spin-constrained DFT formalism with linear constraints is proposed. In this method, on-site magnetic moments and energy functional are automatically optimized, and magnetic excited states can be achieved easily and be described in the accuracy of $10^{-6}$ Bohr magneton and $10^{-5}$ eV, respectively. As an example, the potential energy surface is obtained for itinerant ferromagnetic Fe as a hyper-surface consisting of longitudinal and transverse spin degrees of freedom. By virtue of the high precision, system’s electronic, magnetic and lattice response to spin fluctuations are illustrated in an antiferromagnetic material CrI$_3$. We also find this method particularly suitable for materials genome project, for it can reduce the magnetic torque calculation by magnitude of $N$ (the number of atoms).

Spin fluctuations possess longitudinal and transverse perturbations of magnetic moments which depart from the ground state magnetic configurations. It plays a key role in determining the equilibrium states and the associated dynamical behavior of magnetic systems, such as the phase diagram, transportation of electron, phonon and magnon, etc. Furthermore, it was found to be strongly connected to a number of exotic phenomena, such as superconductivity [11], quantum critical point [12–15], quantum phase transformation [16], topological magnet [17], quantum anomalous Hall effect [18], multiferroics [19,22], etc. Exploring and understanding the microscopic mechanisms behind is an important task of current condensed matter physics research. To interpret the strong coupling between electron, lattice, and spin in these material systems is essential to reveal the microscopic mechanisms of these exotic states. Since the electronic structure of the ground state of magnetic materials has been well explained by first-principles approaches, it is thus desirable to use similar machinery to address the electronic structures, lattice dynamics, and magnetic interactions involving spin fluctuations.

However, the mostly used density functional theory (DFT) is originally [20] designed to depict ground states instead of excited states. Spin polarized relativistic Korringa-Kohn-Rostoker (KKR) Green function method [21] and atomic spin dynamics [22] can give accurate information with arbitrary rotation of magnetization, while still suffered over the complicated on-site modulations from random fluctuations. Lagrange multipliers were then introduced to constrain magnetic moments when solving Kohn-Sham equation as called constrained DFT (cDFT) [20][28]. These cDFT methods not only need users’ high proficiency in interactively adjusting optimization strategy [29], but also experienced difficulty with convergence in itinerant metallic magnetic materials, where the electrons forming magnetic moments are also participate in the formation of a complex Fermi surface and hop across the lattice [39]. Therefore, errors are unavoidably passed into the calculation of magnetic interaction or magnon spectrum, as the result of the rough precision of magnetic moments and energy. These becomes insurmountable obstacle when facing the challenge of nowadays high-throughput depiction, like searching for magnetic materials with several competing local metastable states, topological merits, or high spin transport efficiency, etc.

The convergence requirement for magnetization and energy implies the need for the optimization of both on-site moments and the energy functional (the Lagrangian). The subtlety of magnetic excitation also suggests the necessity of tuning the constraints on the fly. None of the above is included in previous publications. In this letter, we propose a self-adaptive spin-constrained DFT scheme by optimizing on-site magnetic moments and the electronic energy functional alternately within Lagrangian formalism. We first deal with problems involving particular rotations and then generalize the notion of excitation to random spin fluctuations. We utilize it to obtain the excited potential energy surface (PES) and spin torques of an itinerant-electron material Fe, in order to demonstrate its precise and efficient constraining ability. Furthermore, we apply the method to a two-dimensional magnetic material CrI$_3$, investigating its spin effective Hamiltonian as well as its electronic and phonon structure under magnetic excitation.

The target is to minimize the Kohn-Sham energy functional under the constraint that the magnetic moment of the atoms $M_i(\{\phi_i\})$ is the same as the target value de-
noted by $\mathbf{M}_f^*$. We may formulate the problem as

$$\min_{\{\phi_i\}} E_{\text{KS}}(\{\phi_i\})$$

s.t. $M_f(\{\phi_i\}) = M_f^*$

$$\langle \phi_i | \phi_j \rangle = \delta_{ij}$$

We define the Lagrangian as

$$L(\phi_i, \lambda_f, \eta_i) = E_{\text{KS}}(\{\phi_i\}) - \sum_i \eta_i [\langle \phi_i | \phi_j \rangle - \delta_{ij}] - \sum_f \lambda_f \cdot [M_f(\{\phi_i\}) - M_f^*]$$

The necessary condition for the constrain optimization problem is $\nabla_{\phi_i} L = 0$, $\nabla_{\lambda_f} L = 0$ and $\nabla_{\eta_i} L = 0$. Then we have

$$\frac{\delta E_{\text{KS}}}{\delta \langle \phi_i \rangle} - \sum_f \lambda_f \cdot \frac{\delta M_f}{\delta \langle \phi_i \rangle} = \eta_i |\phi_i\rangle$$

$$M_f(\{\phi_i\}) = M_f^*$$

The atomic magnetic moment is generally defined as $\mathbf{M}_f(\{\phi_i\}) = \text{Tr}(\rho \sigma \hat{W}_f)$, where $\rho$ is the density matrix, $\sigma$ is the Pauli matrix, $\hat{W}_f$ is the pre-defined weight operator for atom $I$. One widely chosen weight operator is $\hat{W}_f = \int dr f(r_{\text{cut}} - |r - r_f|) |r\rangle \langle r|$, which is a real-space sphere with smooth boundary $f$. The preferable weight operator in atomic orbital basis DFT is of a different form such as Mulliken partitioning \[31\]. Eq. 3 now becomes

$$\hat{H}_{\text{KS}} |\phi_i\rangle - \sum_f \lambda_f \cdot \sigma \hat{W}_f |\phi_i\rangle = \eta_i |\phi_i\rangle$$

and the Lagrangian multiplier $\lambda_f$ now acts as a "constraining" field.

At first glance, one can not solve the equations Eq. 3 (Eq. 5) and Eq. 4 together, but we may consider the following relaxed scheme (see Fig. 1): (i) Electronic loop: Given $\lambda_f$, solve Eq. 5 we have a self-consistent solution $\{\phi_i(\lambda_f)\}$ that implicitly depends on $\lambda_f$. Note that they may not satisfy the constraint Eq. 4. (ii) Spin loop: Solve an optimization problem $\min_{\lambda_f} \frac{1}{2} \text{Tr}[H_{\text{KS}}(\{\phi_i(\lambda_f)\}) - M_f^*]^2$, which is equivalent to Eq. 4. Nonlinear conjugate gradient (NCG) method is chosen to solve this optimization problem. Given a updated $\lambda_f^*$, we can solve Eq. 5 in the subspace spanned by the old orbitals $\{\phi_i(\lambda_f)\}$ using diagonalization to obtain a new set of orbitals $\{\phi_i(\lambda_f^*)\}$ and thus the moment $\mathbf{M}(\lambda_f^*)$. Therefore the optimization can keep going until the minimum is achieved. We choose the similar perturbation-like scheme as Ref. \[22\] in the second step and significantly reduces the computational cost compared to diagonalization in the whole basis set. In addition, the spin loop converges rapidly because the function $\mathbf{M}(\{\phi_i(\lambda_f)\}) = \partial^2 L/\partial \lambda_f^2$ can be proved to be almost monotonous according to first-order perturbation theory $L(\phi_i, \lambda_f, \eta_i)$ is a concave function of $\lambda_f$ at 0 K \[27\].

Some details of the second step deserve to be discussed here. The gradient of the object function in NCG can be approximated as

$$\langle M_f - M_f^* \rangle \frac{\partial M_f}{\partial \lambda_f}$$

with the assumption that $\partial M_f(\{\phi_i\})/\partial \lambda_f$, the non-local response to the on-site constraining field, $\approx 0$. $\partial M_f/\partial \lambda_f$ can be then obtained by lower-complexity estimation strategies; In order to achieve better convergence properties, we need to control both $\Delta \mathbf{M} = |\mathbf{M}(\{\phi_i(\lambda_f)\}) - \mathbf{M}_f^*|^2$ and the main diagonal of the gradients $\partial \mathbf{M}_f/\partial \lambda_f$ in the spin loop. We give a gradually tightened criterion $\varepsilon_{\text{loc}}$ for the former one, preventing the constraining field from an early-stage local minimum. For the latter one, the loop stops when it is smaller than a empirical value $\varepsilon_{\text{loc}} (\varepsilon_{\text{loc}} \approx 1 \mu_B/\text{eV} \text{ works well across our limited tests})$. This criterion, based on the aforementioned localization assumption, turns out to be extremely crucial.

One can also obtain the magnetic effective field effortlessly based on the Hellmann–Feynman theorem upon the convergence, as

$$B^\text{eff}_I = -\frac{\delta L(M_f^*)}{\delta M_f^*} = -\lambda_f$$

and thus the magnetic torque $\omega_f = -|M_f^*| \mathbf{I} \lambda_f^* M_f^*$, where $\mathbf{I} \lambda_f^* M_f^*$ represents the component perpendicular to $M_f$. It
Two DOFs we scan are the magnitude of atomic magnetic loop (See our analysis in supplementary materials) and may collapse in the early stage of electronic iterations. We found that the energy-favored magnitude increases when spins head towards ferromagnetic (FM) order and decreases when they head towards anti-ferromagnetic (AFM) order. The magnitude-optimized energy curve is actually the PES calculated by former direction-only constraining method, which is a subset of our \( V(\mathbf{M}, \theta) \) PES.

**Effects on magnetic interactions** One of our advantages is to obtain the real “on-site” magnetic interaction around an arbitrary magnetic configuration, by simply calculating the first derivative of energy \( E \) with respect to \( \mathbf{M} \), which is not realistic in previous study because the requirement for precision for both \( E \) and \( \delta \mathbf{M} \) were hard to achieve. In Fig. 3 we show the calculated \( J_{ij} \) and \( K_{ij} \) as the bilinear exchange and biquadratic exchange parameters with the variation of the magnetic configurations. The effective Hamiltonian we utilized is \( H = -\sum_{i,j > i} J_{ij} (\mathbf{e}_i \cdot \mathbf{e}_j) - \sum_{i,j > i} K_{ij} (\mathbf{e}_i \cdot \mathbf{e}_j)^2 \), where \( i, j \) are for atom indices. Both \( J_1 \) and \( K_1 \) exhibit a strong dependency on local magnetic configurations, shown in Fig. 3. \( J_1 \) from previous energy-mapping strategy, which is a constant around 3 meV [33], is somewhere in between the maximum and the minimum of our result [35].

**Effects on lattice dynamics** It is interesting to find out that variation of spin configuration has non-trivial influence on phonon behaviors, and we provide a straightforward way to identify these couplings. This is carried out by combining frozen phonon method [36] together with DeltaSpin. Particularly, one need to calculate the system’s energy precisely at atomic and magnetic configurations, the root-mean-square error (RMSE) between the obtained and the desired spin configuration gets stuck around \( 10^{-2} \mu_B \) (blue lines in Fig. 2(b)). By comparison, DeltaSpin algorithm exhibits a much better convergence, where the RMSE decreases rapidly to zero \( (10^{-8} \mu_B) \) in the same amount of electronic iterations. We found that the energy-favored magnitude increases when spins head towards ferromagnetic (FM) order and decreases when they head towards anti-ferromagnetic (AFM) order. The magnitude-optimized energy curve is actually the PES calculated by former direction-only constraining method, which is a subset of our \( V(\mathbf{M}, \theta) \) PES.

**Potential Energy Surface** A fine-grid potential energy surface (PES) is of great significance especially the one considering spin degree of freedom (DOF). While the previous quadratic cDFT needs a pre-determined multiplier \( \lambda \) and may collapse in the early stage of electronic loop (See our analysis in supplementary materials), our method can self-adaptively update the multiplier and shows a much better precision and efficiency. We study the magnetic PES of BCC iron (See Fig. 2c). Two DOFs we scan are the magnitude of atomic moments \( |\mathbf{M}| \) and the included angle between nearest neighbors’ moments \( \theta \). In calculations using former quadratic constraints, the root-mean-square error (RMSE) between the obtained and the desired spin configuration gets stuck around \( 10^{-2} \mu_B \) (blue lines in Fig. 2(b)). By comparison, DeltaSpin algorithm exhibits a much better convergence, where the RMSE decreases rapidly to zero \( (10^{-8} \mu_B) \) in the same amount of electronic iterations. We found that the energy-favored magnitude increases when spins head towards ferromagnetic (FM) order and decreases when they head towards anti-ferromagnetic (AFM) order. The magnitude-optimized energy curve is actually the PES calculated by former direction-only constraining method, which is a subset of our \( V(\mathbf{M}, \theta) \) PES.

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natic excitation, and at the same time prevent the magnetic configuration from “drifting away”. DeltaSpin can limit the energy and the on-site moment error down to $10^{-9}$ eV and $10^{-7} \mu_B$ respectively yet remains efficient with more than one hundred atoms, and to our knowledge it is the only method that access this type of phonon plots. Four selected spin fluctuated states for CrI$_3$ are obtained (See Fig.4). Notice that only the moments of chromium are constrained while iodine atoms are fully relaxed, thanks to DeltaSpin’s functionality to selectively constrain atoms or components. We found five modes that are significantly influenced by magnetic excitation, which all appears in the high frequency branches where the vibrations of Cr atoms dominate, shown in Fig. 4(a). Regardless of spin-orbit coupling and anti-symmetric exchange, these frequency shifts can be roughly explained by the change of Heisenberg-exchange-contributed force constant $[\partial J_{ij}/\partial r] [e_i \cdot e_j]$. While the second term obviously depends on the spin configuration, the first term, mainly contributed by the competition between the AFM $t_{2g}-t_{2g}$ and FM $t_{2g}-e_g$ interaction [37], also changes remarkably due to the different occupancy of $e_g$ and $t_{2g}$ orbitals across all four configurations. The existence of “collective-motion” modes, in which the distance between any two Cr atoms remains unchanged (Fig.4(b), $\Gamma_2 + \Gamma_3$), suggests that the interaction between Cr and I atoms, i.e. metal-ligand interaction, are also strongly affected by only changing the on-site moments of metal atoms. The fact that the moments of I relax from $10^{-1}\mu_B$ to nearly zero as $\theta$ goes from $0^\circ$ to $180^\circ$ agrees with the magnetic charge theorem [38, 39] and supports this inference. Equipped by this algorithm, not only can we obtain the spin-lattice interaction information, but also one can easily start from the objective of obtaining particular magnetic configuration, and find a feasible way of exciting it selectively by lattice vibrations. This can guide the ultrafast THz experiments through resonant excitation of infra-red (IR) [40] or Raman-active phonon [41].

Effects on Electronic Structure Spin fluctuations has prominent effect on electronic structure, such as the band structure and the orbital character of the Fermi surface [12-43]. First-principles calculation of electronic structure with spin fluctuations is a critical input for evaluating many-body effect in dynamic mean field theory (DMFT) [43]. Here we demonstrate the band structure is profoundly affected by spin fluctuations using
our constrained method (See Fig. 3). We construct a number of spin-fluctuated configurations near monolayer CrI$_3$’s ground state, in which spins are drawn at random from the uniform distribution within FM order $\pm 10^\circ$. Their "excited" K-S orbitals are then precisely calculated following DFT routine, which distinguishes our method from TDDFT utilizing the expansion of the ground state. Different branches from the obtained bands show slightly different broadening behavior with amplitude varies around 30-50 meV, and consequently the band gap shift. In addition, we obtain several electronic states with different canting angles $\theta$ in the transition from FM phase to Néel AFM phase of CrI$_3$, which exhibit prominent variation in band gap and Fermi surface orbital features (See supplementary material Fig. 2). This method can be a potentially useful strategy to access spin renormalized band structure that fully considers the coupling between spin and electronic degrees of freedom [3], which proves exceptionally fruitful in investigating fundamental low energy physics or exploring rich functionalities in dynamical or optical properties.

In this paper, we provide a self-adaptive spin-constrained DFT method in which the spin fluctuations or magnetic excitation can be well incorporated with spin-orbital coupling and non-collinear magnetic configurations, with full degree of freedom on amplitude and rotation angle. It equips us the capability to gain an insight into spin fluctuations from electron, lattice and magnetic point of view with high precision of energy, atomic force and magnetic torque. Also, its high speed of calculating energy and the corresponding derivatives make it stand out as a systematic data generator for machine learning surrogate models [40,48] and will be a game changer with first-principles precision and high efficiency.

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