

Self-consistent versus non-self-consistent spin-spiral calculations of magnetic exchange interactions in VASP

Umit Daglum ^{1,*}, Maria Stamenova ¹, Ersoy Şaşıoğlu ², and Stefano Sanvito ¹

¹*School of Physics and CRANN, Trinity College, Dublin 2, Ireland*

²*Institute of Physics and Halle-Berlin-Regensburg Cluster of Excellence CCE, Martin Luther University Halle-Wittenberg, 06120 Halle (Saale), Germany*



(Received 30 October 2025; accepted 1 May 2026; published 1 June 2026)

We present an *ab initio* investigation of magnetic exchange interactions using the spin-spiral method as implemented in the VASP code, comparing fully self-consistent (SC) total-energy calculations with non-self-consistent (NSC) band-energy evaluations within the same computational framework. Using representative 3d ferromagnets (Fe, Co, Ni) and Mn-based full Heusler compounds, we compute magnon dispersion relations and extract real-space Heisenberg exchange parameters from the Fourier transformation of spin-spiral energies. Curie temperatures are subsequently estimated within the mean-field and random-phase approximations. The SC approach yields exchange parameters and magnon spectra in excellent agreement with experimental and previous theoretical data, confirming its quantitative reliability across different classes of magnetic systems. In contrast, the NSC approach based on a frozen-potential approximation exhibits systematic quantitative deviations: it overestimates spin-spiral energies and exchange couplings in high-moment systems (bcc Fe and Mn-based Heuslers) while underestimating them in low-moment fcc Ni. The magnitude of these discrepancies increases with the magnetic moment size and can exceed several hundred percent in high-moment compounds. Our results demonstrate that while NSC evaluations are computationally efficient, fully SC spin-spiral calculations are essential for obtaining quantitatively reliable exchange interactions within the projector augmented-wave framework.

DOI: [10.1103/pfn2-p6jb](https://doi.org/10.1103/pfn2-p6jb)

I. INTRODUCTION

Understanding magnetic interactions in solids is essential for predicting and engineering the properties of a broad range of materials, from elemental ferromagnets to complex spintronic compounds. Central to this understanding are exchange interactions, which govern the collective behavior of localized and itinerant spins and determine key physical quantities such as Curie temperatures, magnon dispersion relations, and the stability of magnetic ground states. Within an effective Heisenberg description of spin excitations, the exchange parameters J_{ij} provide a quantitative measure of the strength and range of spin-spin interactions. These parameters are widely employed in atomistic spin-dynamics simulations and multiscale modeling of magnetic phenomena [1–4].

First-principles calculations based on density functional theory (DFT) have become indispensable for estimating magnetic exchange interactions in real materials. Most computational approaches rely on the adiabatic approximation, in which electronic degrees of freedom are assumed to ad-

just instantaneously to slowly varying spin configurations. Within this framework, several methods have been developed to extract the exchange parameters J_{ij} , including the total-energy mapping technique [5], the Green's function-based Liechtenstein–Katsnelson–Antropov–Gubanov formalism [6–8], and the frozen-magnon or spin-spiral method [9]. The latter is particularly well suited to electronic-structure methods based on Hamiltonian diagonalization, such as the augmented spherical wave [10,11] and linear muffin-tin orbital (LMTO) methods [12]. It has also been implemented in plane-wave electronic-structure codes such as FLEUR [13] and VASP [14,15], where spin spirals are treated within the noncollinear projector augmented-wave (PAW) formalism [16,17]. These adiabatic approaches are computationally efficient and have been successfully applied to a wide range of magnetic materials. However, they neglect dynamical many-body effects, such as Stoner excitations, which can instead be captured by more advanced but computationally demanding techniques such as time-dependent DFT (TDDFT) [18] and many-body perturbation theory (MBPT) [19].

Among the various adiabatic DFT-based techniques, the spin-spiral method provides an efficient framework for describing long-range magnetic interactions and extracting exchange parameters. By exploiting the generalized Bloch theorem, this approach allows noncollinear spin modulations to be imposed within the primitive unit cell, thereby avoiding the large supercells typically required for describing long-wavelength magnetic excitations. This

*Contact author: dalummm@tcd.ie

formalism is particularly advantageous for metallic magnets, where magnetic order arises from itinerant or weakly localized electrons, as in bcc Fe and many Heusler compounds. Spin-spiral calculations, therefore, offer a powerful tool for mapping the magnetic energy landscape and characterizing exchange interactions in complex magnetic materials.

In practice, the energies of spin-spiral states can be evaluated using different computational strategies within the noncollinear PAW framework. One approach is the fully self-consistent (SC) method, in which the electronic charge density and exchange-correlation (XC) potential are fully relaxed for each spiral configuration (within a constrained spin DFT framework). Alternatively, a computationally cheaper but approximate strategy involves a non-self-consistent (NSC) evaluation of band energies. In this procedure, often based on a frozen-potential approximation, the change in the sum of occupied eigenvalues is evaluated starting from a converged ferromagnetic (FM) ground state. While the NSC approach can significantly reduce the computational cost of spin-spiral calculations, its quantitative reliability may be limited by the absence of electronic relaxation, particularly in systems where rotations of the magnetic moments induce non-negligible changes in the exchange-correlation field.

The VASP code provides native support for spin-spiral calculations based on the generalized Bloch theorem within the noncollinear PAW formalism [16,17]. In this work, we perform a systematic comparison of SC total-energy calculations and NSC band-energy evaluations within this framework. Using representative 3d ferromagnets (Fe, Co, Ni) together with Mn-based full Heusler compounds as benchmark systems, we compute magnon dispersion relations, extract Heisenberg exchange parameters, and estimate Curie temperatures within both the mean-field and random-phase approximations (RPAs). Our analysis shows that the SC approach yields exchange parameters and magnon spectra in excellent agreement with previous theoretical and experimental studies, confirming its quantitative reliability across different classes of magnetic systems. In contrast, the NSC approach, which neglects the relaxation of the exchange-correlation potential, leads to systematic deviations whose magnitude increases with the magnetic moment size.

The primary objective of this work is therefore to provide a methodological, code-specific benchmark of spin-spiral calculations as implemented in VASP. By comparing fully SC total-energy calculations with NSC band-energy evaluations within the same computational framework, we assess the quantitative reliability and limitations of these two computational strategies. The materials considered here are deliberately chosen as well-established benchmark systems, enabling a transparent methodological comparison rather than a reevaluation of their previously reported magnetic properties.

The remainder of this paper is organized as follows. Section II presents the formalism for calculating Heisenberg exchange parameters, magnon dispersion relations, and Curie temperatures. Section III describes the computational setup. Section IV contains the results and discussion, including a comparative analysis of spin-spiral energies obtained from the SC and NSC approaches and an examination of the physical

origins of the discrepancies between them. Finally, Sec. V summarizes the main conclusions of this work.

II. THEORETICAL FRAMEWORK

The accurate determination of magnetic exchange interactions is essential for understanding and predicting the thermodynamic and spin-dynamical behavior of magnetic materials. In this section, we present the theoretical framework for computing magnetic interactions using the spin-spiral method within DFT. Within the adiabatic approximation, transverse spin rotations around a magnetic reference state can be described by an effective Heisenberg-like Hamiltonian that captures low-energy spin-wave excitations. We first introduce the spin-spiral formalism used for this purpose and then outline two complementary computational approaches: a SC treatment and a computationally efficient NSC evaluation based on the frozen-potential approximation. Finally, we describe how Curie temperatures can be estimated from the resulting magnon spectra.

While the formalism presented here primarily applies to single-sublattice ferromagnetic systems, such as the elemental 3d ferromagnets (Fe, Co, Ni) and certain Mn-based Heusler compounds, we also consider Heusler materials with multiple magnetic sublattices. For these systems, we employ the generalized spin-spiral formalism for multisublattice magnets [20] to extract sublattice-resolved exchange parameters. Correspondingly, Curie temperatures for these compounds are estimated using the multisublattice mean-field approximation (MFA). Our RPA analysis is restricted to single-sublattice materials where the standard formalism is directly applicable; extensions of the RPA to multisublattice magnets can be found in the literature [21,22].

A. Spin-spiral formalism

Within the adiabatic approximation, transverse spin excitations around a magnetic reference state can be described by an effective Heisenberg-like Hamiltonian that captures the low-energy magnetic interactions,

$$H = - \sum_{i \neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where $\mathbf{S}_i$ are unit vectors describing the orientation of the magnetic moments at lattice site i , and J_{ij} denotes the exchange interaction between sites i and j . In this work, the Heisenberg Hamiltonian is employed solely as an effective adiabatic representation of transverse spin fluctuations. It does not imply the presence of rigid localized moments nor a state-independent description valid at all energy scales. The underlying assumption of the adiabatic approximation is that the electronic degrees of freedom relax much faster than the orientations of the magnetic moments, thereby defining a magnetic energy landscape for slowly varying spin configurations.

In practice, the spin-spiral formalism employed here is based on the generalized Bloch theorem [9], which allows noncollinear magnetic structures with a well-defined spiral wave vector $\mathbf{q}$ to be treated within the primitive unit cell. In this approach, a lattice translation is combined with a

corresponding rotation of the spin quantization axis, leaving the Kohn–Sham Hamiltonian invariant under the combined operation. Consequently, long-wavelength magnetic textures, such as helical spin spirals, can be described without constructing large real-space supercells. In the present work, we employ the noncollinear PAW implementation of spin spirals available in VASP [16,17].

To evaluate J_{ij} from first principles, we employ spin-spiral configurations characterized by a wave vector $\mathbf{q}$ and a cone angle θ :

$$\mathbf{S}_i = (\sin \theta \cos(\mathbf{q} \cdot \mathbf{R}_i), \sin \theta \sin(\mathbf{q} \cdot \mathbf{R}_i), \cos \theta), \quad (2)$$

describing a helical spin texture.

The total energy difference between a spin-spiral state and the ferromagnetic reference ($\mathbf{q} = 0$) is expressed in terms of the Fourier transform of the exchange interactions:

$$\Delta E(\mathbf{q}, \theta) = \sin^2 \theta [J(\mathbf{0}) - \text{Re}J(\mathbf{q})], \quad (3)$$

with

$$J(\mathbf{q}) = \sum_j J_{0j} e^{-i\mathbf{q} \cdot \mathbf{R}_j}, \quad (4)$$

where $J(\mathbf{q})$ is the lattice Fourier transform of the real-space exchange parameters.

The magnon dispersion relation then reads

$$\omega(\mathbf{q}) = \frac{2}{M} [J(\mathbf{0}) - \text{Re}J(\mathbf{q})], \quad (5)$$

where M is the atomic magnetic moment (in μ_B) and $\omega(\mathbf{q})$ is the spin-wave energy at wave vector $\mathbf{q}$.

The inverse Fourier transform provides access to the real-space exchange couplings:

$$J_{0j} = \frac{1}{N \sin^2 \theta} \sum_{\mathbf{q}} \Delta E(\mathbf{q}, \theta) e^{i\mathbf{q} \cdot \mathbf{R}_j}, \quad (6)$$

where N is the number of $\mathbf{q}$ points in the Brillouin zone. This method enables the extraction of Heisenberg exchange parameters directly from first-principles total energy calculations.

B. Estimation of Curie temperature

Once the exchange parameters J_{ij} are determined, T_c can be estimated using either the MFA or the more accurate RPA.

In the MFA, T_c is given by

$$k_B T_c^{\text{MFA}} = \frac{2}{3} \sum_j J_{0j} = \frac{2}{3} J(\mathbf{0}), \quad (7)$$

where k_B is Boltzmann's constant. Although MFA tends to overestimate T_c due to its neglect of spin-wave excitations, it often yields reasonable results in three-dimensional systems with close-packed lattices (e.g., fcc lattice) and long-range exchange couplings.

A more reliable estimate is provided by the RPA, which accounts for the full magnon spectrum [10]:

$$k_B T_c^{\text{RPA}} = \frac{2M}{3\mu_B} \left(\frac{1}{N} \sum_{\mathbf{q}} \frac{1}{\omega(\mathbf{q})} \right)^{-1}, \quad (8)$$

where $\omega(\mathbf{q})$ is the magnon energy obtained from spin-spiral calculations. The RPA captures collective spin-wave effects.

We note that Monte Carlo simulations provide an alternative and widely used route to estimate T_c , typically yielding values close to those from RPA [23]. However, for consistency with the spin-spiral framework, we restrict our analysis to MFA and RPA in this work.

C. Non-self-consistent band-energy approach

A computationally efficient alternative to SC spin-spiral calculations is the NSC evaluation of the magnetic energy landscape. In this approach, the energy of a spin spiral is approximated by the changes in the single-particle band energy, using a frozen-potential approximation, constructed from the FM ground state. Within this framework, the change in energy is estimated from the variation of the sum of occupied Kohn–Sham eigenvalues,

$$\Delta E(\mathbf{q}, \theta) \approx \sum_{\mathbf{k}, n} f(\epsilon_{\mathbf{k}, n}) [\epsilon_{\mathbf{k}, n}(\mathbf{q}, \theta) - \epsilon_{\mathbf{k}, n}(\mathbf{0}, \theta)], \quad (9)$$

where $\epsilon_{\mathbf{k}, n}(\mathbf{q}, \theta)$ and $\epsilon_{\mathbf{k}, n}(\mathbf{0}, \theta)$ denote the Kohn–Sham eigenvalues for the spiral and FM configurations, respectively, and $f(\epsilon_{\mathbf{k}, n})$ is the Fermi–Dirac occupation function.

This NSC treatment substantially reduces the computational cost by avoiding a full self-consistency cycle for each spiral wave vector $\mathbf{q}$. However, it neglects the relaxation of the charge density and exchange–correlation potential in response to the noncollinear spin perturbation. In practical calculations this frozen-potential approximation may therefore lead to quantitative deviations from fully SC results, particularly in systems where spin rotations induce significant changes in the electronic structure.

It is useful to distinguish the present energy-based evaluation from linear-response formulations based on transverse susceptibilities or Green's-function approaches [6,24], which provide an alternative route to exchange interactions and are not considered in the present benchmark.

III. COMPUTATIONAL DETAILS

All calculations were performed using VASP, employing the PAW method [25]. To facilitate a meaningful comparison with available literature results, we used different exchange–correlation functionals for different material classes. For the elemental 3d ferromagnets—bcc Fe, fcc Co, and fcc Ni—we adopted the local density approximation (LDA) [26], which is widely used in previous studies for computing magnon dispersions, exchange parameters, and Curie temperatures in these systems. In contrast, for the Heusler compounds, we employed the generalized gradient approximation in the Perdew–Burke–Ernzerhof (PBE) formulation [27], consistent with the majority of prior first-principles investigations of this material class [28].

Experimental lattice constants were used in all calculations: 2.87 Å for Fe, 3.55 Å for Co, and 3.52 Å for Ni. For the Heusler compounds, we likewise used experimental lattice parameters reported in the literature [29,30]. Brillouin zone integrations were carried out using a Γ -centered $16 \times 16 \times 16$ k -point mesh for SC electronic structure

calculations. Spin-spiral calculations were performed using the generalized Bloch theorem on a uniform $10 \times 10 \times 10$ grid of spiral wave vectors $\mathbf{q}$, with a fixed cone angle of $\theta = 30^\circ$. All other computational parameters, including plane-wave energy cutoff (500 eV), convergence thresholds (10^{-6} eV), and smearing method, were kept consistent across both material classes to ensure comparability of spin-spiral energies and derived magnetic properties.

For the SC approach, we employed the constrained-spin formalism in VASP to maintain the ideal helical configuration. Local constraining fields, introduced via Lagrange multipliers (the penalty functional method), were applied to preserve the prescribed cone angle θ and the directions of the magnetic moments. This ensures that the electronic density and potential are fully relaxed under the constraint of the fixed spin-spiral geometry, providing the reference total energy $E(\mathbf{q})$. For the NSC approach, the band energies were evaluated in a single step using the frozen potential from the converged ferromagnetic ground state, as described in Sec. II. Additional analyses of the penalty energy and moment stability are provided in the Supplemental Material [31].

IV. RESULTS AND DISCUSSION

A. Accuracy of self-consistent and non-self-consistent spin-spiral calculations

The NSC evaluation of spin-spiral energies is often used to estimate magnetic excitation spectra from a fixed ground-state electronic structure, thereby avoiding computationally demanding SC total-energy calculations for each magnetic configuration. This approach relies on a frozen-potential approximation derived from the ferromagnetic ground state and has historically been applied to compute spin-wave dispersions and extract Heisenberg exchange parameters in several electronic-structure frameworks. Nevertheless, recent methodological studies have shown that its quantitative accuracy can depend sensitively on the treatment of the effective potential and on the degree of itinerancy of the magnetic system.

To assess the reliability of the NSC approach for spin-spiral calculations within the VASP PAW framework, we compare the resulting spin-spiral energy dispersions with those obtained from fully SC calculations. As benchmark systems, we first consider the elemental 3d ferromagnets—bcc Fe, fcc Co, and fcc Ni—which represent prototypical itinerant magnets. The goal is to determine whether the NSC band-energy evaluation provides an accurate description of the magnetic energy landscape and the corresponding exchange interactions in these well-studied materials. Unlike linear-response or susceptibility-based formulations associated with the magnetic force theorem, the present comparison specifically tests the validity of neglecting the SC relaxation of the exchange–correlation potential in the presence of finite-angle spin-spiral perturbations.

In the NSC evaluation, spin-spiral energies in Eq. (9) were obtained non-self-consistently using a frozen-potential approximation derived from the ferromagnetic ground state, where the magnetic structure is perturbed by a transverse spiral with wave vector $\mathbf{q}$. For comparison, the SC spin-

spiral calculations were performed with the magnetization constrained to an analogous spiral configuration at a fixed cone angle θ using the penalty functional method. This direct comparison allows us to assess the accuracy of the frozen-potential approximation across a series of ferromagnets with increasing magnetic moment, ranging from Ni to Fe.

Figure 1(a) shows the spin-spiral energy as a function of $\sin^2 \theta$ at $\mathbf{q} = (0, 0, 1)$. The SC results exhibit the linear dependence expected for transverse spin rotations within the adiabatic description across all three materials. The NSC results also display an approximately linear trend but differ significantly in magnitude from the SC reference. For bcc Fe, the NSC approach overestimates the spin-spiral energy by more than a factor of 2 at $\sin^2 \theta = 1$; for fcc Co, the overestimation is about 25%, while for fcc Ni, the NSC evaluation underestimates the energy by roughly 15%. These results indicate that the frozen-potential approximation does not provide quantitatively accurate spin-spiral energies even for elemental ferromagnets.

This behavior is further illustrated in Fig. 1(b), which presents the spin-spiral dispersion $E(\mathbf{q})$ along the high-symmetry Γ –N direction for bcc Fe and Γ –X for fcc Co and Ni, calculated at a fixed cone angle of $\theta = 30^\circ$. For Fe and Co, the NSC evaluation systematically overestimates the spin-spiral energies throughout the Brillouin zone, with the discrepancy increasing toward the zone boundary. In Ni, the NSC and SC results are similar near Γ , but the difference grows with increasing $|\mathbf{q}|$ and reaches roughly 30% at the X point. The consistent deviations observed across these materials suggest that they originate from the neglect of electronic and potential relaxation in the frozen-potential approximation.

The observed discrepancies can be traced to the limitations of the frozen-potential approximation underlying the NSC evaluation. In NSC spin-spiral calculations the directions of the magnetic moments are rotated throughout the unit cell, including the PAW augmentation spheres. At the same time, the XC potential obtained for the ferromagnetic ground state is kept frozen, i.e., the charge density and the magnitude of the XC field are not updated self-consistently, while only the orientation of its magnetic component is rotated according to the imposed spin-spiral configuration. Within the PAW formalism, the XC field extends over the entire unit cell, and a substantial fraction of it resides in the interstitial region shared by several atoms. As a result, the rotation of the magnetic moments induces changes in the magnetic energy that are not properly captured within the frozen-potential approximation, and the interstitial part of the XC field may contribute to the observed band-energy variations. Similar behavior has been reported previously in FLAPW-based spin-spiral studies by Ležaić *et al.* [20]. In those calculations, quantitative agreement between NSC and SC spin-spiral energies was obtained only after removing the interstitial contribution to the XC field in the frozen-potential evaluation, whereas retaining this contribution led to a substantial overestimation of the spin-spiral energies. Although a similar trend is observed here, the PAW formalism does not allow for a direct separation of interstitial contributions as in FLAPW, and therefore the underlying mechanism may not be strictly identical. A related tendency for frozen-potential approaches to overestimate magnon

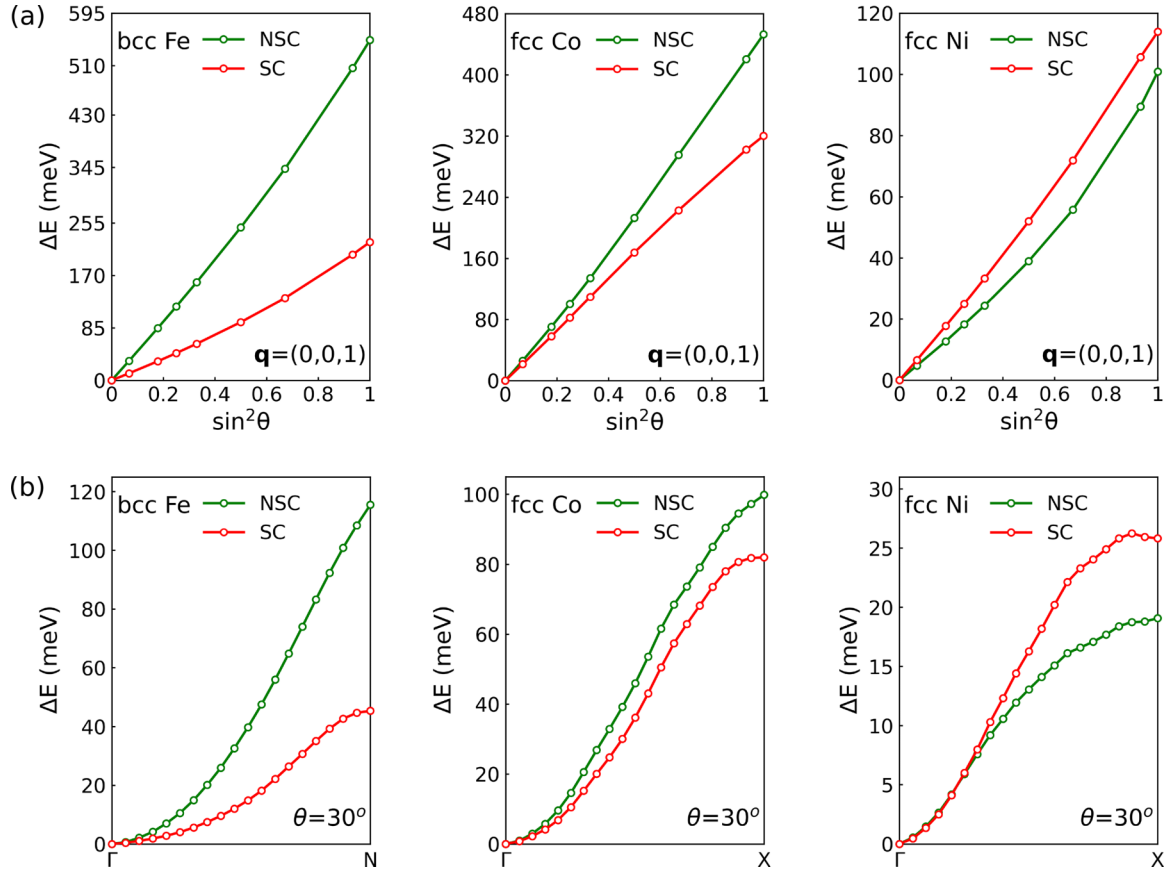


FIG. 1. Comparison of spin-spiral energies obtained from non-self-consistent (NSC) band-energy evaluations and fully self-consistent (SC) total-energy calculations for bcc Fe, fcc Co, and fcc Ni. (a) Spin-spiral energy as a function of $\sin^2 \theta$ at the wave vector $\mathbf{q} = (0, 0, 1)$. Both approaches exhibit the linear dependence expected within the adiabatic description of transverse spin rotations, while the NSC results show noticeable quantitative deviations from the SC reference, particularly for bcc Fe. (b) Spin-spiral dispersion $E(\mathbf{q})$ along the high-symmetry Γ -N direction for bcc Fe and Γ -X for fcc Co and fcc Ni, calculated at a fixed cone angle of $\theta = 30^\circ$. The NSC calculations systematically deviate from the SC results, overestimating the exchange energies in Fe and Co and slightly underestimating them in Ni near the Brillouin-zone boundary.

energies has also been reported recently by dos Santos *et al.* [32], who observed comparable overestimations of magnon energies derived from frozen-potential exchange parameters in NiO and MnO.

These considerations indicate that the NSC frozen-potential evaluation may not always provide quantitatively reliable spin-spiral energies within the VASP PAW framework. The issue becomes particularly pronounced in systems with multiple magnetic sublattices, where the interstitial part of the XC potential contributes substantially to the magnetic energy landscape. For this reason, the remaining analysis presented below is based on fully SC spin-spiral calculations. As demonstrated in the following sections, the SC approach yields magnon dispersions and Heisenberg exchange parameters in good agreement with available experimental data and previous theoretical studies.

B. Spin-wave dispersion, exchange interactions, and Curie temperatures in Fe, Co, and Ni

Having established the quantitative limitations of the frozen-potential approximation within the VASP framework, we now focus on the SC spin-spiral calculations

to investigate spin-wave excitations and magnetic interactions in elemental 3d ferromagnets. The systems considered here—bcc Fe, fcc Co, and fcc Ni—represent a progression from strong to weak ferromagnetism and from more localized to more itinerant magnetic character. They therefore provide well-established benchmark systems for analyzing spin dynamics and exchange interactions and allow us to assess the ability of the SC approach to capture the magnetic energy landscape of prototypical itinerant magnets.

Before discussing the calculated dispersions, it is useful to briefly recall the two principal types of spin excitations in metallic ferromagnets: collective spin-wave (magnon) modes and single-particle spin-flip (Stoner) excitations. Spin-wave excitations correspond to coherent transverse oscillations of the magnetization and are often well described by effective Heisenberg models, where exchange interactions are obtained from total-energy differences or linear-response approaches. In contrast, Stoner excitations arise from transitions between spin-split electronic bands and represent incoherent single-particle processes. Their proper description requires a dynamical treatment of the transverse magnetic susceptibility, as provided by TDDFT or MBPT, which also capture magnon

damping and finite lifetimes [18,19]. Inelastic neutron scattering experiments typically reveal both types of excitations, although well-defined spin-wave modes dominate the low-energy region.

Since the present spin-spiral calculations are based on static total-energy differences, they provide direct access only to the coherent spin-wave dispersion and do not include lifetime effects associated with coupling to Stoner excitations. Consequently, the approach does not describe the damping or broadening of spin-wave modes that may occur at larger wave vectors in metallic systems. Nevertheless, it provides a reliable description of the energy of coherent magnons and can be directly compared with experimental measurements and with TDDFT or MBPT calculations in the low-energy regime, where damping effects remain relatively small.

Figure 2 presents the spin-wave dispersions obtained from the SC spin-spiral calculations, compared with the frozen-potential results of Halilov *et al.* [12] and the dynamical TDDFT calculations of Buczek *et al.* [33]. For all three materials, the SC dispersions closely follow the TDDFT spectra in both shape and magnitude, supporting the reliability of the SC total-energy approach for describing the adiabatic energy landscape of low-energy spin-wave excitations. In contrast, the frozen-potential results of Halilov *et al.* systematically underestimate the spin-wave energies, with deviations becoming more pronounced from bcc Fe to fcc Ni. This trend correlates with the decreasing magnetic moment and increasing itinerant character of the ferromagnetic state, highlighting the limitations of neglecting the SC electronic response. As emphasized by Bruno [34], frozen-potential approximations tend to lose accuracy in materials with smaller magnetic moments because they do not account for the SC reconfiguration of the exchange-correlation field under noncollinear perturbations. The close agreement between the present SC results and the TDDFT data therefore indicates that static total-energy spin-spiral calculations provide a reliable description of coherent magnon energies, particularly in the low- to intermediate- $\mathbf{q}$ region where these modes remain well-defined.

To further analyze the low-energy behavior of the computed spin-wave spectra, we extract the spin-wave stiffness constants D by fitting the curvature of the SC dispersion near the Γ point. These values, presented in Table I, reflect the increasing rigidity of the spin system from Fe to Ni, with D rising from 220 meVÅ² in bcc Fe to 544 meVÅ² in fcc Co and reaching 757 meVÅ² in fcc Ni. The results for Fe and Co are in good agreement with experimental data and prior theoretical estimates, demonstrating the accuracy of the SC approach in these itinerant ferromagnets. In fcc Ni, however, the calculated D substantially overestimates the experimental range. This discrepancy highlights the breakdown of the adiabatic Heisenberg mapping in the weak-moment, strongly itinerant regime. In this limit, the separation between collective magnons and single-particle Stoner excitations becomes less distinct, and hybridization effects—captured only by more advanced dynamical frameworks such as MBPT [19]—play a decisive role in renormalizing the spin-wave energies. While such dynamical renormalization and damping effects lie beyond the scope of static total-energy calculations, the extracted stiffness constants provide a consistent SC benchmark

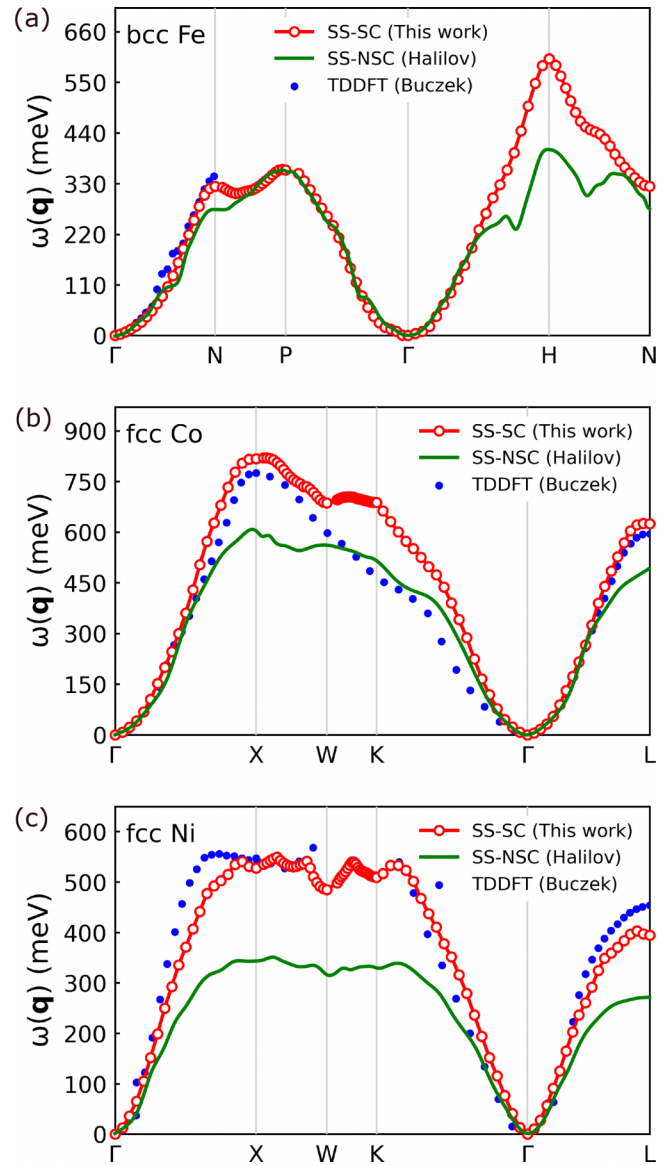


FIG. 2. Spin-wave dispersions along high-symmetry directions of the Brillouin zone for (a) bcc Fe, (b) fcc Co, and (c) fcc Ni. Red circles denote the fully self-consistent spin-spiral (SC) calculations obtained in this work. The results are compared with the frozen-potential calculations of Halilov *et al.* [12] (solid green lines) and the time-dependent density functional theory (TDDFT) results of Buczek *et al.* [33] (blue circles). The close agreement between the present SC results and the TDDFT data in the low- and intermediate-energy range indicates that the spin-spiral approach provides an accurate description of coherent spin-wave excitations in these itinerant ferromagnets.

for the adiabatic energy landscape in the long-wavelength limit.

While the spin-wave stiffness constant D characterizes the magnetic rigidity in the long-wavelength limit, a microscopic understanding of the magnetic interactions requires the extraction of real-space exchange couplings. To this end, we obtain the Heisenberg exchange parameters J_{ij} by performing an inverse Fourier transform of the SC spin-spiral energy dispersion $E(\mathbf{q})$. In this framework, the spatial resolution

TABLE I. Calculated and experimental magnetic properties of bcc Fe, fcc Co, and fcc Ni: lattice constants (a), magnetic moments (m), Curie temperatures (T_C) from MFA and RPA, and spin-wave stiffness constants (D).

	a (Å)	m (μ_B)	T_C^{MFA} (K)	T_C^{RPA} (K)	$T_C^{\text{expt.}}$ (K)	$D_{\text{calc.}}$ (meVÅ ²)	$D_{\text{expt.}}$ (meVÅ ²)
bcc Fe	2.87	2.25	1194	820	1043	220	314 ^c
			1077 ^a	950 ^b		250 ^b	280 ^d
			1414 ^b				
fcc Co	3.55	1.61	1711	1383	1388	544	510 ^e
			1645 ^b	1311 ^b		663 ^b	580 ^f
fcc Ni	3.52	0.60	579	530	627	757	422 ^f
			533 ^a	350 ^b		756 ^b	550 ^g
			397 ^b				

^aReference [35].

^bReference [36].

^cReference [37].

^dReference [38].

^eReference [39].

^fReference [40].

^gReference [41].

and the maximum interaction range are determined by the density of the $\mathbf{q}$ -point mesh used for the Brillouin zone sampling. Our chosen mesh allows for the resolution of exchange interactions up to a distance of $R \approx 3.5a$ (where a is the lattice constant), ensuring that both the dominant short-range couplings and the decaying long-range contributions characteristic of metallic systems are captured. This real-space representation facilitates a direct comparison with previous studies employing alternative electronic structure methodologies.

Figure 3 presents the calculated Heisenberg exchange parameters J_{ij} for bcc Fe, fcc Co, and fcc Ni as a function of the interatomic distance R/a . In all three materials, the magnetic stability is governed by the nearest-neighbor (NN) interaction, which reaches approximately 21 meV in Fe, 18 meV in Co, and 6 meV in Ni. These prominent NN values stem from the strong direct overlap of the partially filled 3d orbitals. The second-nearest-neighbor (2NN) couplings are significantly weaker—approximately 5 meV for Fe, nearly vanishing for Co, and slightly antiferromagnetic (≈ -1 meV) for Ni. For larger interatomic separations, the J_{ij} values exhibit the characteristic oscillatory behavior associated with Ruderman-Kittel-Kasuya-Yosida (RKKY)-like interactions, reflecting the itinerant nature of the magnetism and the presence of a well-defined Fermi surface in these metallic ferromagnets.

A noticeable difference appears in the spatial profile of the exchange couplings among the three systems. In bcc Fe, the J_{ij} parameters extend over longer distances and exhibit more pronounced oscillations, which is consistent with its classification as a weak ferromagnet. In such systems, majority-spin 3d states partially cross the Fermi level, enabling magnetic interactions to be mediated by itinerant s electrons and partially delocalized 3d states through an RKKY-like mechanism. In contrast, fcc Co and especially fcc Ni exhibit stronger ferromagnetism, with fully occupied majority-spin 3d bands and

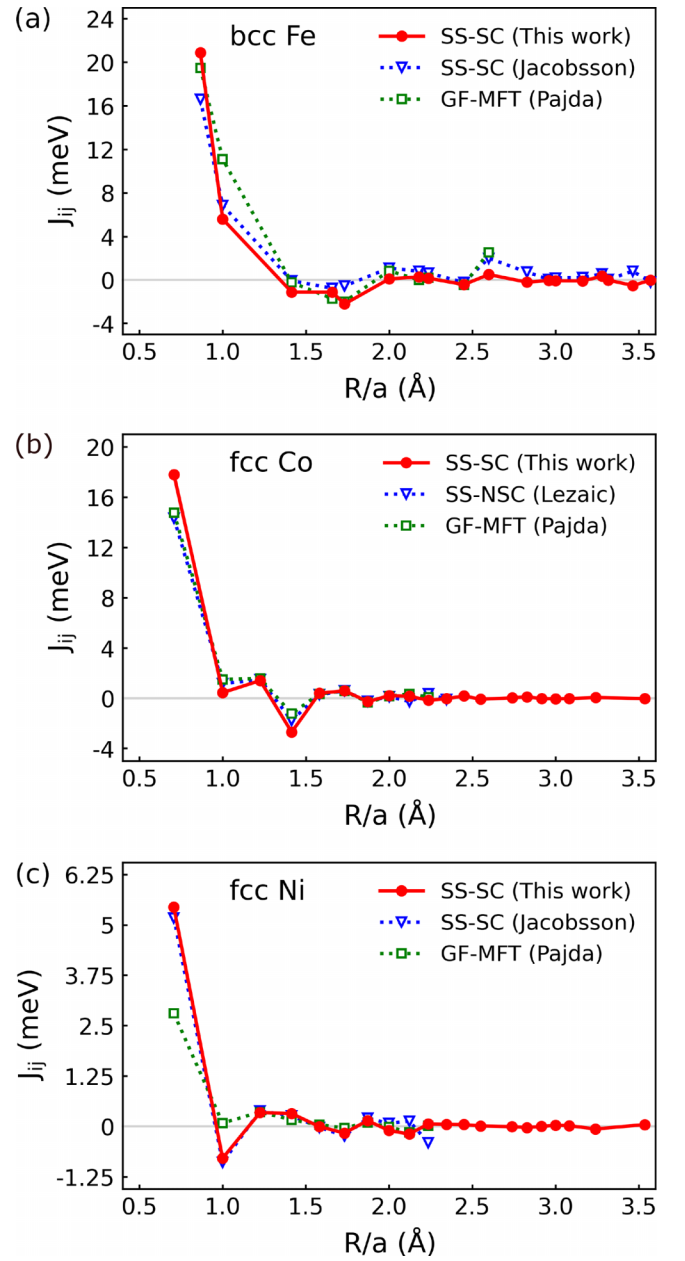


FIG. 3. Heisenberg exchange parameters J_{ij} as a function of interatomic distance R/a (where a is the lattice constant) for (a) bcc Fe, (b) fcc Co, and (c) fcc Ni. The results obtained from the constrained self-consistent (SC) spin-spiral calculations in this work are compared with previous studies: Pajda *et al.* [36], based on the real-space magnetic force theorem within the TB-LMTO method; Jacobsson *et al.* [35], using fully self-consistent spin-spiral calculations with the ELK code; and Ležaić *et al.* [20], employing a frozen-potential spin-spiral evaluation within the FLAPW framework. The close agreement between the present VASP-based SC results and the self-consistent ELK data demonstrates the consistency of the self-consistent spin-spiral approach across different electronic-structure implementations.

a lower density of states at the Fermi level, resulting in more rapidly decaying and short-ranged exchange interactions.

Compared to the real-space results of Pajda *et al.* [36], our SC spin-spiral results systematically yield larger

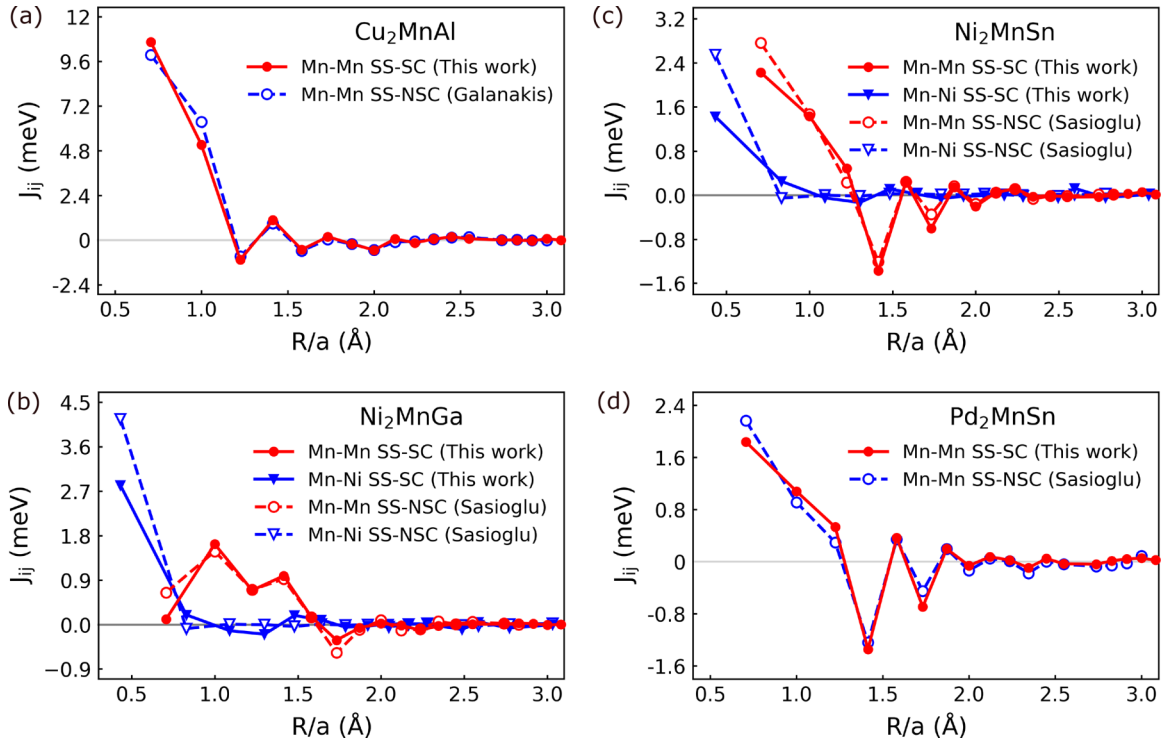


FIG. 4. Heisenberg exchange parameters J_{ij} as a function of interatomic distance R/a (where a is the lattice constant) for the Mn-based full Heusler compounds (a) Cu_2MnAl , (b) Ni_2MnGa , (c) Ni_2MnSn , and (d) Pd_2MnSn . All exchange parameters shown here were obtained from constrained self-consistent (SC) spin-spiral calculations. For Cu_2MnAl and Pd_2MnSn , where the magnetic interactions are dominated by Mn–Mn exchange, the results are compared with the calculations of Galanakis *et al.* [29]. For the multisublattice systems Ni_2MnGa and Ni_2MnSn , both Mn–Mn and Mn–Ni exchange interactions are displayed and compared with the results of Şaşıoğlu *et al.* [30].

J_{ij} values. This is consistent with the tendency of frozen-potential approximations to underestimate the energy of magnetic excitations in itinerant systems. The discrepancy is especially pronounced in fcc Ni, where the exchange interactions are significantly renormalized by the SC response of the exchange-correlation field. Our J_{ij} profiles for Fe and Ni show excellent agreement with the SC results of Jacobsson *et al.* [35], who employed the SC approach within the ELK code. For Co, our results are in good agreement with the data of Ležaić *et al.* [20], while also remaining consistent with the trends observed by Pajda. These comparisons reinforce the reliability and transferability of our SC total-energy approach for extracting real-space exchange parameters across different 3d ferromagnetic environments.

To further validate the precision of the SC approach within the PAW framework, we benchmark our results against all-electron calculations performed with the FLEUR code. For bcc Fe and fcc Ni, our J_{ij} values show excellent agreement with the SC spin-spiral results of Jacobsson *et al.* [35]; notably in Ni, both the interaction magnitude and the spatial decay profile are nearly identical. For fcc Co, our results follow the qualitative trends reported by Ležaić *et al.* [20], though our SC values exhibit systematically higher magnitudes. This difference again reflects the underestimation intrinsic to the frozen-potential approximation used in the latter study, even within an all-electron FLAPW basis (see Fig. 3). It is significant that the data from Ležaić and Pajda for Co align closely with each other, reinforcing the consistency between

real-space and reciprocal-space frozen-potential implementations. These comparisons demonstrate that our SC approach in VASP effectively captures the exchange parameters with close to all-electron-level accuracy, providing a robust foundation for studying magnetic interactions in itinerant ferromagnets.

Finally, we estimate the Curie temperatures (T_C) of bcc Fe, fcc Co, and fcc Ni using both the MFA and the more accurate RPA, based on the extracted Heisenberg exchange parameters. The results are listed in Table I, together with experimental values and previous theoretical estimates. As expected, MFA systematically overestimates T_C due to its neglect of spin-wave fluctuations, while RPA generally provides improved agreement with experiment. For fcc Co and Ni, the RPA results are in good agreement with experimental values: 1383 K versus 1388 K for Co, and 530 K versus 627 K for Ni, significantly outperforming the corresponding MFA estimates. For bcc Fe, however, the RPA value of 820 K underestimates the experimental T_C of 1043 K and is the lowest among all available theoretical predictions, including prior RPA-based spin-spiral estimates in the range of 950–1000 K [36].

C. Exchange interactions and Curie temperatures in Mn-based full Heusler compounds

Heusler compounds provide a versatile platform for exploring diverse magnetic exchange mechanisms due to their chemical tunability and the presence of multiple, often inequivalent, magnetic sublattices [42]. Depending on their composition and the specific occupancy of the X , Y , and

Z sites, these materials can host various magnetic phases, including ferromagnetic, ferrimagnetic, and noncollinear orders. In systems with multiple magnetic sublattices, such as Co_2MnSi , the magnetic ground state is shaped by a complex interplay between intra- and intersublattice couplings [43]. By contrast, many Mn-based full Heuslers ($X_2\text{MnZ}$) contain a single magnetic Mn sublattice, where exchange is typically dominated by long-range interactions mediated by the conduction electron gas.

Given the diversity of exchange mechanisms in these systems, we have also assessed the validity of the frozen-potential approximation in Mn-based Heusler compounds. As detailed in the Supplemental Material [31], a direct comparison between SC and NSC spin-spiral energies reveals that the NSC approach can lead to drastic discrepancies in multisublattice systems. For example, in Pd_2MnSn , the NSC evaluation overestimates the spin-spiral energies by nearly 380 % compared to the SC reference. This breakdown highlights the fundamental difficulty of partitioning the exchange–correlation potential when multiple magnetic species contribute to the interstitial magnetization. These findings confirm that a full treatment of electronic relaxation is essential for complex magnetic structures. Consequently, all Heisenberg exchange parameters reported in this section are obtained exclusively from fully SC spin-spiral calculations. Having established the robustness of the SC approach, we now examine its predictive accuracy for four representative Mn-based full Heusler compounds: Cu_2MnAl , Ni_2MnSn , Pd_2MnSn , and Ni_2MnGa presented in Fig. 4. These materials provide stringent benchmark systems due to the availability of high-quality experimental measurements and detailed theoretical studies, particularly those of Şaşıoğlu *et al.* [47]. To place the present results in context, it is useful to contrast their exchange mechanisms with those of the elemental $3d$ ferromagnets discussed in the previous section. In bcc Fe, fcc Co, and fcc Ni, magnetic stability is primarily governed by strong nearest-neighbor exchange interactions arising from the direct overlap of $3d$ orbitals. As a result, the corresponding J_{ij} profiles are dominated by short-range couplings, while more distant interactions are strongly suppressed and survive only as weak oscillations mediated by the conduction (sp) electrons.

The four Mn-based Heusler compounds studied here share a common structural feature: a single Mn atom per unit cell and Mn–Mn distances exceeding 4 Å. These large separations result in negligible direct $3d$ – $3d$ orbital overlap, rendering direct Mn–Mn exchange inefficient. As a consequence, magnetic interactions are primarily mediated by conduction electrons through indirect mechanisms such as RKKY-type exchange or antiferromagnetic superexchange. Among the compounds considered, Cu_2MnAl exhibits the strongest nearest-neighbor Mn–Mn exchange interaction (~ 11 meV) and a sizable second-nearest-neighbor coupling (~ 5 meV), with subsequent couplings showing an oscillatory decay.

In Ni_2MnGa , the first three Mn–Mn interactions are ferromagnetic, although the nearest-neighbor term is nearly negligible. This compound also features a strong nearest-neighbor Mn–Ni exchange of ~ 4 meV, enabled by short Mn–Ni separation and $3d$ – $3d$ hybridization, despite the small Ni magnetic moment ($\sim 0.3 \mu_B$). Ni_2MnSn and Pd_2MnSn , being isoelec-

TABLE II. Lattice constants (a), atom-resolved spin magnetic moments (m_X and m_Y), total magnetic moments, and Curie temperatures for the studied Mn-based full Heusler compounds. Both mean-field (MFA) and random phase approximation (RPA) estimates are reported alongside available experimental data. For comparison, selected results from previous theoretical studies are also included.

Material $X_{(2)}YZ$	a (Å)	m_X (μ_B)	m_Y (μ_B)	m_{total} (μ_B)	T_c^{MFA} (K)	T_c^{RPA} (K)	T_c^{Exp} (K)
Cu_2MnAl	5.95	0.075 0.02 ^b	3.55 3.67 ^b	3.56 3.60 ^b	1008 970 ^b	713 635 ^b	603 ^a
Pd_2MnSn	6.38	0.07 0.07 ^b	3.97 4.08 ^b	4.11 4.16 ^b	275 252 ^b	242 178 ^b	189 ^c
Ni_2MnGa	5.85	0.33 0.29 ^e	3.44 3.57 ^e	4.03 4.09 ^e	402 389 ^e		380 ^d
Ni_2MnSn	5.99	0.23 0.21 ^e	3.53 3.72 ^e	3.92 4.08 ^e	397 358 ^e		360 ^d

^aReference [44].

^bReference [29].

^cReference [45].

^dReference [46].

^eReference [30].

tronic and structurally similar, exhibit nearly identical J_{ij} profiles: the first three Mn–Mn exchange parameters are ferromagnetic, as expected from the empirical Castelliz-Kanomata argument [48,49], followed by long-range oscillatory couplings. Minor differences stem from variations in their lattice constants (see Table II). In Ni_2MnSn , a nearest-neighbor Mn–Ni exchange interaction is also observed, though it is smaller in magnitude than the corresponding Mn–Mn interaction.

Across all four compounds, the calculated exchange parameters show excellent agreement with previous results, particularly those reported by Galanakis *et al.* [29] and Şaşıoğlu *et al.* [30], thereby validating the accuracy of our SC spin-spiral approach. It is worth noting that the large magnetic moments characteristic of Mn-based Heuslers lead to negligible differences between NSC and fully SC spin-spiral calculations, as also confirmed in Ref. [30]. This agreement forms a solid foundation for evaluating the magnetic ordering tendencies and Curie temperatures in the subsequent analysis.

The Curie temperatures computed from the extracted Heisenberg exchange parameters are summarized in Table II, alongside experimental values and results from previous theoretical studies. For all four compounds, we report the MFA estimates of T_c , which provide a convenient upper bound but are known to overestimate the transition temperature due to their neglect of collective spin fluctuations. To obtain more realistic predictions, we also evaluate T_c within the RPA, which incorporates spin-wave excitations. However, our RPA implementation is currently limited to systems with a single magnetic sublattice, and hence RPA estimates are only provided for Cu_2MnAl and Pd_2MnSn . For these two compounds, the RPA T_c values are in excellent agreement with previous MFT-based studies and show a marked improvement over MFA in terms of quantitative agreement with experimental data. Specifically, for Cu_2MnAl we find $T_c^{\text{RPA}} = 713$ K, close to the experimental value of 603 K, while Pd_2MnSn yields $T_c^{\text{RPA}} = 242$ K compared to the experimental 189 K. In the case of Ni_2MnSn

and Ni_2MnGa , which contain both Mn and Ni magnetic atoms, RPA calculations would require a full multisublattice formalism. We therefore restrict our analysis to the MFA values for these compounds, which are found to be in good agreement with previous theoretical work and within 10% of the experimental values. Overall, the computed T_c values corroborate the trends observed in the exchange interactions and further confirm the reliability of the SC spin-spiral approach for estimating finite-temperature magnetic properties.

Finally, we note that the quantitative accuracy of spin-spiral calculations is influenced not only by the degree of self-consistency but also by the underlying exchange-correlation functional. Standard local and semilocal functionals such as LDA and PBE are well-known to overestimate the exchange splitting in itinerant ferromagnets, most notably in fcc Ni, where both the magnitude and anisotropy of the splitting can deviate substantially from experiment [19]. This intrinsic limitation affects the magnetic excitation spectrum and the extracted exchange parameters largely independently of whether SC or NSC spin-spiral approaches are employed. In this context, recently developed exactly constrained $U(1)\times\text{SU}(2)$ -symmetric exchange-correlation functionals within Spin-Current Density Functional Theory provide a formally rigorous framework for noncollinear magnetism. These approaches, which require full self-consistency by construction, offer a consistent treatment of spin currents and exchange-correlation torques, and thus represent a promising direction for improving the quantitative reliability of spin-spiral calculations in weak and strongly itinerant magnetic systems [50–52].

V. SUMMARY AND CONCLUSIONS

In this work, we have carried out a systematic first-principles investigation of magnetic exchange interactions using the spin-spiral method within the VASP code, comparing fully SC total-energy calculations with NSC frozen-potential band-energy evaluations. The aim of this study is methodological: to assess the quantitative reliability of these two computational strategies for evaluating spin-spiral energies within the same electronic-structure framework. Our analysis considered two representative classes of magnetic systems, namely, elemental $3d$ ferromagnets (bcc Fe, fcc Co, and fcc Ni) and Mn-based full Heusler compounds (Cu_2MnAl , Ni_2MnSn , Pd_2MnSn , and Ni_2MnGa). For each material, we calculated spin-spiral energies, extracted real-space Heisenberg exchange parameters J_{ij} through inverse Fourier transformation, and estimated Curie temperatures within both the MFA and RPA.

The fully self-consistent spin-spiral approach yields magnon dispersions and exchange parameters in very good agreement with previous theoretical studies and available experimental data, demonstrating its quantitative reliability across different magnetic environments. In contrast, the NSC frozen-potential evaluation produces systematic deviations. It tends to underestimate spin-wave energies in weak-moment itinerant systems such as fcc Ni, while significantly overestimating them in systems with larger and more localized magnetic moments. This behavior is particularly pronounced in the Mn-based full Heusler compounds, where deviations from the SC results can reach several hundred percent. These discrepancies originate from the neglect of charge-density and exchange-correlation-field relaxation in the frozen-potential approximation when the magnetic moments are rotated.

Overall, our results indicate that fully self-consistent spin-spiral calculations are required for obtaining quantitatively reliable exchange parameters and magnon spectra within the VASP PAW framework. The systematic deviations observed in the NSC approach, ranging from moderate errors in low-moment systems to severe overestimations in high-moment magnets, illustrate the limitations of the frozen-potential approximation for describing the magnetic energy landscape of itinerant and multisublattice systems. The present study therefore provides a methodological benchmark and practical guidance for future first-principles investigations of magnetic interactions and spin excitations in complex materials.

ACKNOWLEDGMENTS

This work was supported by Research Ireland under Grant No. 18/SIRG/5515. The authors acknowledge the DJEI/DES/SFI/HEA, Trinity College Dublin Research IT and the Irish Center for High-End Computing (ICHEC) for providing computational resources. U.D. gratefully acknowledges M. Schlipf for insightful discussions and valuable feedback. E.Ş. acknowledges financial support from the Deutsche Forschungsgemeinschaft (DFG) through the Collaborative Research Center CRC/TRR 227.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

- [1] W. Heisenberg, Zur Theorie des Ferromagnetismus, *Z. Phys.* **49**, 619 (1928).
- [2] V. P. Antropov, M. I. Katsnelson, B. N. Harmon, M. Van Schilfgaarde, and D. Kusnezov, Spin dynamics in magnets: Equation of motion and finite temperature effects, *Phys. Rev. B* **54**, 1019 (1996).

- [3] P.-W. Ma, C. H. Woo, and S. L. Dudarev, Large-scale simulation of the spin-lattice dynamics in ferromagnetic iron, *Phys. Rev. B* **78**, 024434 (2008).
- [4] A. V. Ruban, S. Shallcross, S. I. Simak, and H. L. Skriver, Atomic and magnetic configurational energetics by the generalized perturbation method, *Phys. Rev. B* **70**, 125115 (2004).

- [5] T. Archer, C. D. Pemmaraju, S. Sanvito, C. Franchini, J. He, A. Filippetti, P. Delugas, D. Puggioni, V. Fiorentini, R. Tiwari, *et al.*, Exchange interactions and magnetic phases of transition metal oxides: Benchmarking advanced *ab initio* methods, *Phys. Rev. B* **84**, 115114 (2011).
- [6] A. I. Liechtenstein, M. Katsnelson, V. Antropov, and V. Gubanov, Local spin density functional approach to the theory of exchange interactions in ferromagnetic metals and alloys, *J. Magn. Magn. Mater.* **67**, 65 (1987).
- [7] X. Wan, Q. Yin, and S. Y. Savrasov, Calculation of magnetic exchange interactions in Mott-Hubbard systems, *Phys. Rev. Lett.* **97**, 266403 (2006).
- [8] H. J. Xiang, E. J. Kan, S.-H. Wei, M.-H. Whangbo, and X. G. Gong, Predicting the spin-lattice order of frustrated systems from first principles, *Phys. Rev. B* **84**, 224429 (2011).
- [9] L. Sandratskii, Noncollinear magnetism in itinerant-electron systems: Theory and applications, *Adv. Phys.* **47**, 91 (1998).
- [10] L. M. Sandratskii and P. Bruno, Exchange interactions and Curie temperature in (Ga,Mn)As, *Phys. Rev. B* **66**, 134435 (2002).
- [11] J. Kübler, *Ab initio* estimates of the Curie temperature for magnetic compounds, *J. Phys.: Condens. Matter* **18**, 9795 (2006).
- [12] S. V. Halilov, H. Eschrig, A. Y. Perlov, and P. M. Oppeneer, Adiabatic spin dynamics from spin-density-functional theory: Application to Fe, Co, and Ni, *Phys. Rev. B* **58**, 293 (1998).
- [13] The FLEUR project, <https://www.flapw.de/>.
- [14] G. Kresse and J. Hafner, *Ab initio* molecular dynamics for liquid metals, *Phys. Rev. B* **47**, 558 (1993).
- [15] G. Kresse and J. Furthmüller, Efficiency of *ab-initio* total energy calculations for metals and semiconductors using a plane-wave basis set, *Comput. Mater. Sci.* **6**, 15 (1996).
- [16] D. Hobbs, G. Kresse, and J. Hafner, Fully unconstrained noncollinear magnetism within the projector augmented-wave method, *Phys. Rev. B* **62**, 11556 (2000).
- [17] M. Marsman and J. Hafner, Broken symmetries in the crystalline and magnetic structures of γ -iron, *Phys. Rev. B* **66**, 224409 (2002).
- [18] S. Y. Savrasov, Linear response calculations of spin fluctuations, *Phys. Rev. Lett.* **81**, 2570 (1998).
- [19] E. Şaşıoğlu, A. Schindlmayr, C. Friedrich, F. Freimuth, and S. Blügel, Wannier-function approach to spin excitations in solids, *Phys. Rev. B* **81**, 054434 (2010).
- [20] M. Ležaić, P. Mavropoulos, G. Bihlmayer, and S. Blügel, Exchange interactions and local-moment fluctuation corrections in ferromagnets at finite temperatures based on noncollinear density-functional calculations, *Phys. Rev. B* **88**, 134403 (2013).
- [21] E. Şaşıoğlu, L. Sandratskii, P. Bruno, *et al.*, Magnetic exchange coupling and Curie temperature of $\text{Ni}_{(1+x)}\text{MnSb}$ ($x = 0, 0.25, 0.5, 0.75, 1$) from first principles, *J. Magn. Magn. Mater.* **290**, 385 (2005).
- [22] J. Rusz, I. Turek, and M. Diviš, Random-phase approximation for critical temperatures of collinear magnets with multiple sublattices: GdX compounds ($X = \text{Mg, Rh, Ni, Pd}$), *Phys. Rev. B* **71**, 174408 (2005).
- [23] K. Binder and D. W. Heermann, *Monte Carlo Simulation in Statistical Physics* (Springer, 1992), Vol. 8.
- [24] F. L. Durrhus, T. Skovhus, and T. Olsen, Plane wave implementation of the magnetic force theorem for magnetic exchange constants: Application to bulk Fe, Co and Ni, *J. Phys.: Condens. Matter* **35**, 105802 (2023).
- [25] P. E. Blöchl, Projector augmented-wave method, *Phys. Rev. B* **50**, 17953 (1994).
- [26] J. P. Perdew and Y. Wang, Accurate and simple analytic representation of the electron-gas correlation energy, *Phys. Rev. B* **45**, 13244 (1992).
- [27] J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized gradient approximation made simple, *Phys. Rev. Lett.* **77**, 3865 (1996).
- [28] S. Sanvito, C. Oses, J. Xue, A. Tiwari, M. Zic, T. Archer, P. Tozman, M. Venkatesan, M. Coey, and S. Curtarolo, Accelerated discovery of new magnets in the Heusler alloy family, *Sci. Adv.* **3**, e1602241 (2017).
- [29] I. Galanakis and E. Şaşıoğlu, *Ab-initio* calculation of effective exchange interactions, spin waves, and Curie temperature in L_{21} - and L_{12} -type local moment ferromagnets, *J. Mater. Sci.* **47**, 7678 (2012).
- [30] E. Şaşıoğlu, L. M. Sandratskii, and P. Bruno, First-principles calculation of the intersublattice exchange interactions and Curie temperatures of the full Heusler alloys Ni_2MnX ($X = \text{Ga, In, Sn, Sb}$), *Phys. Rev. B* **70**, 024427 (2004).
- [31] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/pfn2-p6jb> for additional computational details, convergence tests, and benchmarking results of the constrained self-consistent spin-spiral method, including analyses of the Wigner-Seitz radius, penalty parameter, and comparisons between NSC and self-consistent spin-spiral calculations.
- [32] F. J. dos Santos, L. Binci, G. Menichetti, R. Mahajan, N. Marzari, and I. Timrov, Comparative study of magnetic exchange parameters and magnon dispersions in NiO and MnO from first principles, *Phys. Rev. B* **113**, 024427 (2026).
- [33] P. Buczek, A. Ernst, and L. M. Sandratskii, Different dimensionality trends in the Landau damping of magnons in iron, cobalt, and nickel: Time-dependent density functional study, *Phys. Rev. B* **84**, 174418 (2011).
- [34] P. Bruno, Exchange interaction parameters and adiabatic spin-wave spectra of ferromagnets: A “renormalized magnetic force theorem,” *Phys. Rev. Lett.* **90**, 087205 (2003).
- [35] A. Jacobsson, G. Johansson, O. I. Gorbatov, M. Ležaić, B. Sanyal, S. Blügel, and C. Etz, Efficient parameterisation of non-collinear energy landscapes in itinerant magnets, *Sci. Rep.* **12**, 18987 (2022).
- [36] M. Pajda, J. Kudrnovský, I. Turek, V. Drchal, and P. Bruno, *Ab initio* calculations of exchange interactions, spin-wave stiffness constants, and Curie temperatures of Fe, Co, and Ni, *Phys. Rev. B* **64**, 174402 (2001).
- [37] M. Stringfellow, Observation of spin-wave renormalization effects in iron and nickel, *J. Phys. C* **1**, 950 (1968).
- [38] J. Lynn, Temperature dependence of the magnetic excitations in iron, *Phys. Rev. B* **11**, 2624 (1975).
- [39] G. Shirane, V. Minkiewicz, and R. Nathans, Spin waves in 3d metals, *J. Appl. Phys.* **39**, 383 (1968).
- [40] R. Pauthenet, Experimental verification of spin-wave theory in high fields, *J. Appl. Phys.* **53**, 8187 (1982).
- [41] P. W. Mitchell and D. M. Paul, Low-temperature spin-wave excitations in nickel, by neutron triple-axis spectroscopy, *Phys. Rev. B* **32**, 3272 (1985).

- [42] T. Graf, C. Felser, and S. S. Parkin, Simple rules for the understanding of Heusler compounds, [Prog. Solid State Chem.](#) **39**, 1 (2011).
- [43] E. Şaşıoğlu, L. M. Sandratskii, P. Bruno, and I. Galanakis, Exchange interactions and temperature dependence of magnetization in half-metallic Heusler alloys, [Phys. Rev. B](#) **72**, 184415 (2005).
- [44] Y. Noda and Y. Ishikawa, Spin waves in Heusler alloys Pd_2MnSn and Ni_2MnSn , [J. Phys. Soc. Jpn.](#) **40**, 690 (1976).
- [45] K. Tajima, Y. Ishikawa, P. J. Webster, M. W. Stringfellow, D. Tocchetti, and K. R. Zeabeck, Spin waves in a Heusler alloy Cu_2MnAl , [J. Phys. Soc. Jpn.](#) **43**, 483 (1977).
- [46] P. J. Webster and K. R. A. Ziebeck, Alloys and compounds of d-elements with main group elements, in *Alloys and Compounds of d-Elements with Main Group Elements*, Landolt-Börnstein, New Series, Group III Vol. 19/c, edited by H. R. J. Wijn (Springer, Berlin, 1988), pp. 2 and 75–184.
- [47] E. Şaşıoğlu, L. M. Sandratskii, and P. Bruno, Role of conduction electrons in mediating exchange interactions in Mn-based Heusler alloys, [Phys. Rev. B](#) **77**, 064417 (2008).
- [48] L. Castelliz, Beitrag zum Ferromagnetismus von Legierungen der übergangsmetalle mit Elementen der B-Gruppe, [Z. Metallk](#) **46**, 198 (1955).
- [49] T. Kanomata, K. Shirakawa, and T. Kaneko, Effect of hydrostatic pressure on the Curie temperature of the Heusler alloys Ni_2Mnz ($z = \text{Al, Ga, In, Sn and Sb}$), [J. Magn. Magn. Mater.](#) **65**, 76 (1987).
- [50] J. K. Desmarais, G. Vignale, K. Bencheikh, A. Erba, and S. Pittalis, Electron localization function for non-collinear spins, [Phys. Rev. Lett.](#) **133**, 136401 (2024).
- [51] J. K. Desmarais, A. Erba, G. Vignale, and S. Pittalis, Meta-generalized-gradient approximation made magnetic, [Phys. Rev. Lett.](#) **134**, 106402 (2025).
- [52] M. Desmarais, G. Vignale, and S. Pittalis, Physical spin torques from exactly constrained exchange-correlation torques, [Phys. Rev. Lett.](#) **136**, 016403 (2026).