Interatomic Exchange Interactions for Finite-Temperature Magnetism and Nonequilibrium Spin Dynamics

A. Szilva,[1] M. Costa,[1,2,3] A. Bergman,[1] L. Szunyogh,[4] L. Nordström,[1] and O. Eriksson[1]

[1]*Department of Physics and Astronomy, Division of Materials Theory, Uppsala University, Box 516, SE-75120 Uppsala, Sweden*
[2]*Instituto de Física, Universidade Federal Fluminense, 24210-346 Niterói, Rio de Janeiro, Brazil*
[3]*Department of Physics and Astronomy, University of California, Irvine, California 92697, USA*
[4]*Department of Theoretical Physics and Condensed Matter Research Group of Hungarian Academy of Sciences,*
Budapest University of Technology and Economics, Budafoki út 8., H1111 Budapest, Hungary

We derive *ab inito* exchange parameters for general noncollinear magnetic configurations, in terms of a multiple scattering formalism. We show that the general exchange formula has an anisotropiclike term even in the absence of spin-orbit coupling, and that this term is large, for instance, for collinear configuration in bcc Fe, whereas for fcc Ni it is quite small. We demonstrate that keeping this term leads to what one should consider a biquadratic effective spin Hamiltonian even in the case of collinear arrangement. In noncollinear systems this term results in new tensor elements that are important for exchange interactions at finite temperatures, but they have less importance at low temperature. To illustrate our results in practice, we calculate for bcc Fe magnon spectra obtained from configuration-dependent exchange parameters, where the configurations are determined by finite-temperature effects. Our theory results in the same quantitative results as the finite-temperature neutron scattering experiments.

The noncollinear magnetic alignment, when a global magnetization axis is not easily identified, can appear as the ground state of several magnetic materials [1], e.g., spin-spiral or spin-glass systems, for nonequilibrium, dynamical systems and for magnets at finite temperatures where it is universal. Considering the rigid spin and adiabatic approximation, the calculation of exchange interaction between well-defined moments is crucial for atomistic, first-principles spin-dynamics simulations and for the interpretations of experimental results. One example of an experimental work where a detailed calculation of the interatomic exchange interaction for different noncollinear spin configurations is needed is the Skyrmion state, e.g., of bulk $Fe_{0.5}Co_{0.5}Si$, where this complex magnetic structure is stabilized at finite temperature [2]. Similar questions arise for the Skyrmion state of thin films, e.g., of FeGe, where finite temperature plays a cruical role in stabilizing a Skyrmion crystal [3,4]. Another example of where little is known in detail about finite-temperature effects of interatomic exchange is the classical Invar alloys, e.g., the Fe-Ni alloy in the fcc lattice, which is known to have a zero thermal expansion which is caused by several competing noncollinear configurations that are tuned by finite temperature and volume [5]. For this material the interatomic exchange clearly is expected to depend strongly on temperature and volume [5], although a detailed knowledge is lacking since theoretical tools directly coupling spin configuration to interatomic exchange are lacking. Materials where the interatomic exchange has an apparent temperature (or strain) driven transition from favoring a collinear arrangement to a more complex structure, like a Skyrmion lattice or spin spiral, is not unique for metals, it can also be seen in insulators like for the doped antiferromagnet $La_2Cu_{0.97}Li_{0.03}O_4$ [6].

Clearly, the electronic structure and, as a direct consequence via the multiple scattering formalism of Ref. [7], the evaluation of the interatomic exchange interactions depend in general on the configuration of the atomic spins. This calls for a self-consistency procedure where, for a starting guess of the spin configuration, the interatomic exchange parameters of a suitable spin Hamiltonian are evaluated, after which a new spin configuration can be determined, e.g., by means of atomistic spin dynamics, and the whole procedure may be iterated until self-consistency. Although the formula in the case of collinear arrangement has been known for a long time, due to the seminal work of Ref. [7], even for correlated systems [8], a counterpart for noncollinear arrangement is lacking. In this Letter we derive a general formula for both collinear and noncollinear spin systems, where we make use of the magnetic force theorem [9,10]. This is combined with spin-dynamics simulations to evaluate relevant spin configurations as a function of finite temperature. Our analysis is expressed in terms of multiple scattering formalism (MSF) [11]. Analyzing the one- and two-site spin rotations, we map the analytically derived exchange parameters onto effective spin Hamiltonians, and discuss their appropriateness for collinear and noncollinear spin arrangements, and we illustrate our results with a numerical simulation of the magnon energies of bcc Fe at elevated temperature. Examples of other direct applications of the method

described here relate to ultrafast spin dynamics in multi-sublattice magnets [12] and to the analysis of the remagnetization process in ultrafast magnetization processes [13].

The fundamental equation of a scalar relativistic MSF is given as [11]

$$(\tau_{ij}^{-1})_{L\sigma,L'\sigma'} = P_{iL\sigma\sigma'}\delta_{ij}\delta_{LL'} - G^0_{ij,LL'}\delta_{\sigma\sigma'}, \quad (1)$$

where τ_{ij} stands for the scattering path operator (SPO), P_i denotes the inverse of the single site scattering operator (ISO), L stands for the angular momentum and magnetic quantum numbers, σ refers to the spin index, G^0 is the free (or bare) structure constant, and indices i and j refer to the considered lattice sites. Later on in our presentation we omit the orbital and spin indices. We introduce a general notation for the ISO as follows:

$$P_i(\varepsilon) = p_i^0 I_2 + \vec{p}_i \cdot \vec{\sigma} = p_i^0 I_2 + p_i \vec{n}_i \cdot \vec{\sigma}, \quad (2)$$

where the unit vector $\vec{n}_i$ refers to the magnetic spin moment at site i, $\vec{\sigma}$ are the Pauli matrices, I_2 is the unit matrix in spin space, p_i^0 denotes the nonmagnetic part, and the vector $\vec{p}_i$ stands for the magnetic part of the ISO. Introducing a similar notation for the SPO, it can be written that

$$\tau_{ij}(\varepsilon) = T^0_{ij} I_2 + \vec{T}_{ij} \cdot \vec{\sigma}, \quad (3)$$

where the vector $\vec{T}_{ij}$ has three (x, y, and z) matrix components, which enables us to treat a noncollinear arrangement. We write the variation of ISO as

$$\delta P_i = p_i \delta\vec{n}_i \cdot \vec{\sigma}, \quad (4)$$

where $\delta\vec{n}_i$ stands for the deviation of a spin moment after an infinitesimal rotation at site i. We also introduce the tensor

$$A_{ij}^{\alpha\beta} = \frac{1}{\pi} \int_{-\infty}^{\varepsilon_F} d\varepsilon \operatorname{Im} \operatorname{Tr}_L(p_i T^\alpha_{ij} p_j T^\beta_{ji}), \quad (5)$$

where indices α and β run over 0, x, y, or z. We note that $A_{ij}^{\alpha\beta} = A_{ji}^{\beta\alpha}$ because of the properties of the trace. The collinear alignment is an important special case when the global coordinate system can be chosen so that the vector $\vec{T}_{ij}$ has only nonzero values of the z component between every site, implying all $A_{ij}^{\alpha\beta}$'s are equal to zero except for A_{ij}^{00} and A_{ij}^{zz}. We henceforth refer to A_{ij}^{00} and A_{ij}^{zz} as the collinear exchange parameters, and the other elements as noncollinear exchange parameters. (The collinear alignment usually corresponds to the ferromagnetic ground state in this work.) Introducing quantities $T^\uparrow_{ij} = T^0_{ij} + T^z_{ij}$ and $T^\downarrow_{ij} = T^0_{ij} - T^z_{ij}$ for collinear systems and using the time-reversal symmetry, one obtains the well-known expression [7]

$$A_{ij}^{00} - A_{ij}^{zz} = \frac{1}{\pi} \int_{-\infty}^{\varepsilon_F} d\varepsilon \operatorname{Im} \operatorname{Tr}_L(p_i T^\uparrow_{ij} p_j T^\downarrow_{ji}), \quad (6)$$

which can be defined for any (noncollinear) configuration as

$$J_{ij}^L = A_{ij}^{00} - A_{ij}^{zz}, \quad (7)$$

and will be referred to as the Liechtenstein-Katsnelson-Antropov-Gubanov (LKAG) formula.

According to Andersen's local force theorem [9,10], the total energy variation can be written as the variation of the integrated density of states time energy (first moment). The so-called Lloyd formula [14] says how to calculate this from ISO and SPO, see Eqs. (2) and (3), in the presence of any perturbation, e.g., for a rotation of a spin moment. We speak of *one-site* spin rotation when this perturbation is given due to a rotated magnetic moment only at one site i with infinitesimal angle $\delta\theta$. In that case the detailed derivation of the total energy variation is written in Appendix A in the Supplemental Material [15]; here, we give the final result in collinear limit ($T^x_{ij} = T^y_{ij} \simeq 0$),

$$\delta E_i^{\text{one}} = -2\sum_{j\neq i} J_{ij}^L \delta n_i^z. \quad (8)$$

Note that this equation is obtained without making any assumption of an effective spin Hamiltonian, and is instead a direct consequence of multiple scattering theory. Therefore, any effective spin Hamiltonian should reproduce the results of Eq. (8), in the collinear limit, as regards the energy of one-site rotations. It should be noted that δn_i^z is proportional to $(\delta\theta)^2$; therefore, $\sum_{j\neq i} J_{ij}^L$ is positive for a ferromagnetic ground state. The effective (Weiss) field can be obtained from Eq. (8), and any desired spin Hamiltonian should recover it in the collinear limit.

Next, we consider *two-site* spin moment rotations; i.e., two spin moments at site i and j are rotated simultaneously in opposite directions with angle $\delta\theta$. Using the Lloyd formula we find that an interaction term appears in the variation of the total energy expression. This variation can be written for the general, noncollinear case as

$$\delta E_{ij}^{\text{two}} = \delta E_{ij}^{\text{HT}} + \delta E_{ij}^{\text{AT}}, \quad (9)$$

where

$$\delta E_{ij}^{\text{HT}} = -2\left(A_{ij}^{00} - \sum_{\mu=x,y,z} A_{ij}^{\mu\mu}\right)\delta\vec{n}_i \cdot \delta\vec{n}_j, \quad (10)$$

and

$$\delta E_{ij}^{\text{AT}} = -4\sum_{\mu,\nu=x,y,z} \delta n_i^\mu A_{ij}^{\mu\nu} \delta n_j^\nu; \quad (11)$$

i.e., we obtain a Heisenberg-type (HT) and an anisotropic-type (AT) term expressed by generalized exchange parameters, as given in Eq. (5). The details of the derivation of Eqs. (9)–(11) can be found in Appendix B in the Supplemental Material [15].

As we mentioned earlier, only parameters A_{ij}^{00} and A_{ij}^{zz} should be considered in the collinear limit; therefore, for this case, the two-site energy variation formula simplifies to

$$\delta E_{ij}^{\text{two}} = -2J_{ij}^{L}(\delta n_i^x \delta n_j^x + \delta n_i^y \delta n_j^y) - 2G_{ij}\delta n_i^z \delta n_j^z, \quad (12)$$

where $G_{ij} = A_{ij}^{00} + A_{ij}^{zz}$, implying that we have to deal with two parameters to describe the exchange interaction even in the case of collinear spin arrangement. The parameter J_{ij}^{L} describes the transversal (x or y) part of the energy variation and the longitudinal (z) part is characterized by the parameter G_{ij}, which are proportional to $(\delta\theta)^2$ and $(\delta\theta)^4$, respectively. In the collinear limit it is sufficient to keep only the HT term in Eq. (9), as was done in Ref. [7]. In this case $\delta E_{ij}^{\text{two}}$ equals $\delta E_{ij}^{\text{HT}}$, which can be briefly written as $-2J_{ij}^{L}\delta\vec{n}_i \cdot \delta\vec{n}_j$. By limiting to the *bilinear scalar* Heisenberg effective spin model with exchange parameter J_{ij}^{L}, we also recover in the collinear limit the energy variation described by Eq. (8), as derived in Appendix C in the Supplemental Material [15], a result that is in agreement with Ref. [7].

However, in order to keep both HT and AT terms in the general two-site MSF energy deviation formula (9), we attempt to map the MSF parameters onto a *bilinear tensorial* effective Hamiltonian; see Eq. (44) in Appendix C in the Supplemental Material [15]. In Appendix C of Ref. [15] we derive the total energy variation from the tensorial spin Hamiltonian and show that it is not consistent with the expression of Eq. (8), a fact that implies that one *cannot* map noncollinear MSF parameters onto a tensorial effective Hamiltonian.

This motivates us to take an alternate approach and consider higher order spin terms in the spin Hamiltonian, in the spirit of Ref. [16]. The simplest extension is the *biquadratic* effective Hamiltonian

$$\mathcal{H}^{Q} = -\sum_{ij}^{i \neq j} J_{ij}'\vec{n}_i \cdot \vec{n}_j - \sum_{ij}^{i \neq j} B_{ij}(\vec{n}_i \cdot \vec{n}_j)^2, \quad (13)$$

where a revised bilinear parameter J_{ij}' is introduced in addition to the biquadratic one B_{ij}. Deriving the two-site energy variation formula from Eq. (13), we obtain that the biquadratic two-site rotation energy deviation can be written as a sum of a biquadratic Heisenberg-type term and anisotropic-type term; see Eqs. (51) and (52) in Appendix C of Ref. [15], respectively.

Comparing Eqs. (10) and (11) with Eqs. (51) and (52) in Appendix C of Ref. [15], one can identify

$$J_{ij}' = A_{ij}^{00} - 3A_{ij}^{zz}, \qquad B_{ij} = A_{ij}^{zz}. \quad (14)$$

In the case of one-site rotations, the leading term can be written as $\delta E_i^{\text{one}} = -2\sum(J_{ji}' + 2B_{ji})\delta n_i^z$, where $J_{ji}' + 2B_{ji} = A_{ji}^{00} - A_{ji}^{zz} = J_{ji}^{L}$, i.e., Eq. (8) has been recovered. This is a required condition for any effective spin Hamiltonian, since the analysis from multiple scattering theory establishes Eq. (8). Hence, the recovery of Eq. (8) when considering one-site rotations shows that the collinear MSF parameters *can* be mapped onto a biquadratic model.

The numerical calculations of these parameters have been implemented in terms of a real-space linear muffin tin orbital atomic sphere approximation code; see Ref. [17]. The linear muffin tin orbital formalism used in this work and its connection to MSF is discussed in Ref. [18], and it has been shown that it results in LKAG parameters which are consistent with other electronic structure methods [19]. The calculated exchange parameters between the first nearest neighbor sites are shown in Figs. 1 and 2 for bcc Fe and fcc Ni, respectively. We obtained that $A_{ij}^{zz} = B_{ij}$ is much larger than A_{ij}^{00} for nearest neighbors in bcc Fe. Also, J_{ij}^{L} is drastically different than J_{ij}', showing the importance of higher order spin interactions; see Fig. 1. It might seem, from Fig. 1, that the biquadratic Hamiltonian (with J_{ij}' and B_{ij}) and the bilinear Hamiltonian (with J_{ij}^{L}) give different excitation energies. In the collinear limit they actually give rise to the same excitation energies, since, as follows from Eqs. (7) and (14), the relationships $J_{ij}' + 2B_{ij} = J_{ij}^{L}$ and, consequently, $\delta E_{ij}^{\text{HT}} = \delta E_{ij}^{\text{QHT}}$ hold for all pairs. On the other hand, in the case of fcc Ni first neighbor pairs, the A_{ij}^{zz} and, therefore, the biquadratic parameter are very small, so that J_{ij}^{L} and J_{ij}' are close to each other, as shown in Fig. 2. Figure 2 shows that A_{ij}^{zz} (hence, B_{ij}) deviates from the general trend, in the case of fourth nearest neighbor interaction. This is counterbalanced by a larger value of J_{ij}' for this interaction distance. Hence, also in this case will the biquadratic Hamiltonian and the bilinear Hamiltonian give rise to the same excitation energies, in the collinear limit. The data in Fig. 2 result in a lower value of B_{ij}, which implies that the bilinear term is more dominating for fcc Ni.

Most importantly, our formulation allows us to consider noncollinear spin configurations and to calculate magnon
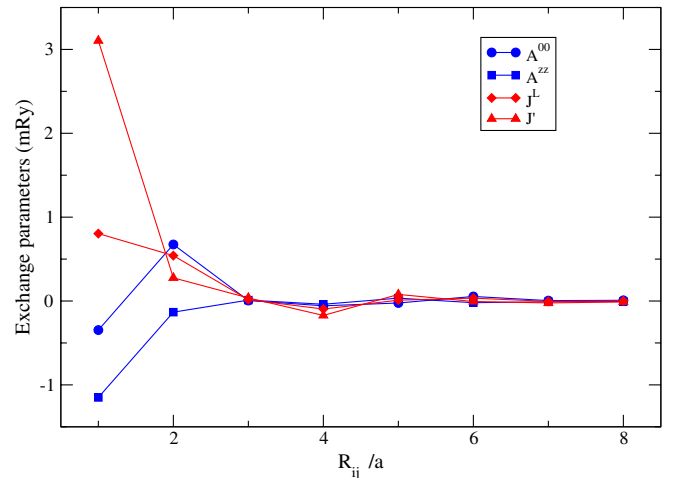

FIG. 1 (color online). Collinear exchange parameters (A^{00} and A^{zz}) between the first eight neighbors in bcc Fe. J^{L}, see Eq. (7), and J', see Eq. (14), are derived parameters for the bilinear and biquadratic spin Hamiltonians, the biquadratic B equals A^{zz}, which is rather large for nearest neighbors.
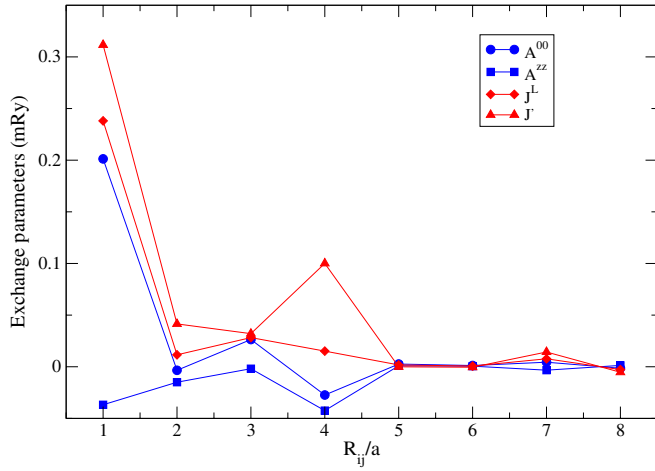
FIG. 2 (color online).   Collinear exchange parameters between
the first eight neighbors in fcc Ni. The bilinear parameter of
bilinear model J^L and the bilinear parameter of biquadratic
model J' are very close to each other in fcc Ni, $A^{zz} \ll A^{00}$.

spectra from these configurations. In Fig. 3 we show the
calculated exchange parameters between two nearest neigh-
bors, when rotating only one spin of a bcc Fe lattice. It can
be seen that the collinear parameters are decreasing and the
noncollinear parameters are increasing when θ increases.

In order to estimate how the finite temperature induced
spin order of the lattice influences the exchange interaction,
and the magnetic excitation energies, we performed
Monte Carlo simulations (with 128 Fe atoms) using the
parameters of Eq. (7), and we performed a statistical analy-
sis of the distribution of angles between the spins on the
simulation box. We find that at 300 K the average deviation
of an atomic spin moment from the global quantization axis
is $\hat{\theta} = 28°$, while in the case of 500 K this deviation was
$\hat{\theta} = 39°$. We then performed a calculation of the
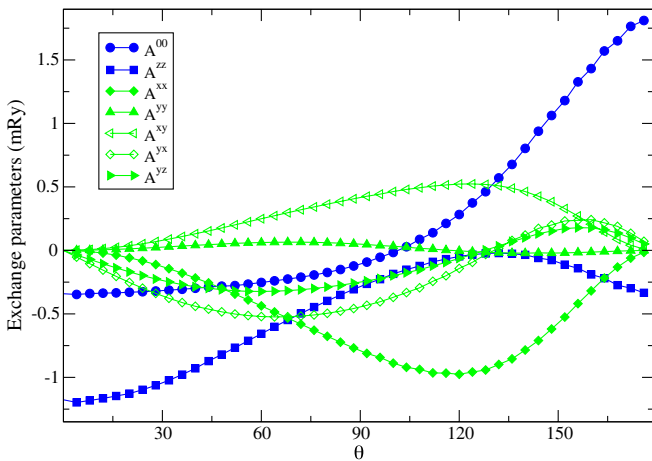

FIG. 3 (color online).   Comparison of the collinear exchange
parameters (blue circles and blue squares) and noncollinear
parameters (green diamonds and green triangles) between two
nearest neighbor sites of bcc Fe as a function of angle rotating
one magnetic moment with angle θ.

parameters in Eqs. (5) and (14) using a spin configuration
with deviations ($\hat{\theta}$'s) from the global magnetization direc-
tion, with angles given by the Monte Carlo simulations
(e.g., 28° at room temperature). Similarly to the case
when only one spin moment was rotated, the noncollinear
parameters become significant for larger average spin-
moment deviation. We then performed a statistical analysis
of these parameters and obtained averages over different
site and μ, ν indices. We analyzed these average exchange
parameters in the cases of $\hat{\theta} = 0°$, 5°, 10°, 25°, and 40°.
For small angles, the collinear parameters are, as expected,
dominant. At room temperature the noncollinear parame-
ters are roughly 30% of the collinear ones, and at 500 K, i.e.,
in the case of 40° average angles between atomic and global
magnetization direction, the collinear and noncollinear pa-
rameters are of the same order.

We then calculated the spin wave spectra along the Γ-H
direction, for finite temperature configurations. The result
is shown in Fig. 4. These spin-wave spectra are obtained
from configuration-dependent LKAG exchange parame-
ters evaluated for noncollinear configurations correspond-
ing to temperatures ranging from 0 to 500 K. As Fig. 1
shows that the first and second neighbor values are domi-
nant, we evaluated the spectra from these parameters only.
At zero temperature ($\theta = 0$) we obtained a spin-wave
stiffness constant of 287 meV Å^2, whereas a magnetization
measurement at 4.2 K resulted in $D_{expt} = 280$–330 meV Å^2
[20,21]. In Fig. 4 we compare our theory with the experi-
mental data of Ref. [20], where the top thick (black) line
shows the calculated spectrum from collinear LKAG ex-
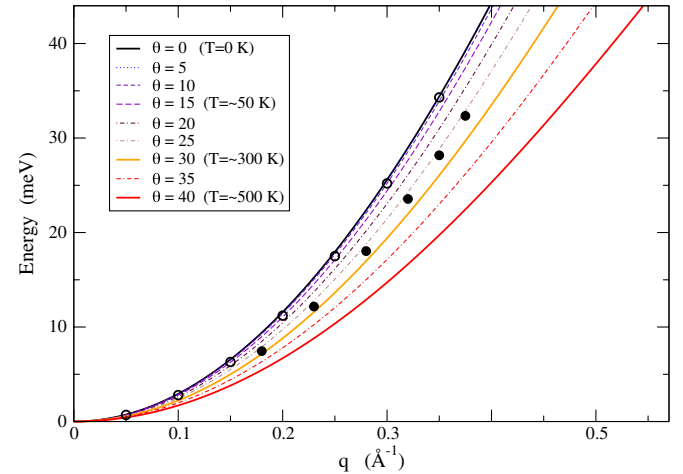change parameters, and the open circles represent

FIG. 4 (color online).   Spin-wave dispersion relation calculated
at different temperatures along the Γ-H direction. The top thick
(black) line shows the calculated spectrum from collinear LKAG
exchange parameters, open circles come from magnetization
measurement at 4.2 K [20] The middle thick (yellow) line
corresponds to the calculated spectrum at 300 K and the filled
circles refer to the room temperature neutron scattering mea-
surement data [22].

experimental data. These experimental values were evaluated from the experimental spin-wave stiffness constant, using the expression $D_{\text{expt}}q^2$, and it may be seen that experiment and theory agree. Reference [22] has carefully examined the temperature dependence of magnetic excitations of iron from neutron scattering data, and the measured room temperature spin-wave spectrum of bcc Fe is shown in Fig. 4 by filled black circles. It is found that experimental values are close to our calculated room temperature (yellow) curve. The experimental room temperate spin stiffness value is 230, to be compared to our calculated value of 219 meV Å^2. Furthermore, Fig. 2. in Ref. [22] shows measured spectra along the (110) direction, starting from low values and increasing T up to the Curie temperature. The measured trend is obvious, softer curves are observed with increasing temperature, in a fashion which is similar to our calculations (Fig. 4).

We have here derived a general expression for the interatomic exchange coupling that describes the interaction between magnetic moments for both collinear and noncollinear (finite temperature) spin configurations. The possibility to evaluate exchange interactions from the electronic structure of a noncollinear spin system allows us to consider an effective spin Hamiltonian with temperature-dependent exchange coupling parameters. It opens up for finite temperature investigations via a self-consistency procedure of both the electronic structure and spin configurations, as described in the introduction of this Letter. This enables accurate finite-temperature spin-dynamics simulations of magnetic materials, and forms a platform for analyzing magnetization dynamics in general, including ultrafast dynamics. The approach described here is aimed at finding the most relevant part of the space of spin configurations, and to evaluate as accurately as possible the interatomic exchange interaction here. This is different from the approach of spin-cluster expansions that aim at introducing sufficiently many terms (of the order of several 30–50) in an expansion of the exchange energy [23], so that a good description of *any* possible configuration could be obtained, albeit without self-consistency of the atomic spin system. Whether cluster expansions, or the approach proposed here, is better suited to in general describe the dynamics of magnetic materials becomes a numerical exercise where the two different methods should be compared in how they reproduce a large body of experimental finite-temperature data. Such an exhaustive comparison is outside the scope of the present study.

[1] J. Kubler, *Theory of Itinerant Electron Magnetism* (Clarendon, Oxford, 2000).

[2] P. Milde, D. Khler, J. Seidel, L. M. Eng, A. Bauer, A. Chacon, J. Kindervater, S. Mhlbauer, C. Pfleiderer, S. Buhrandt, C. Schtte, and A. Rosch, Science **340**, 1076 (2013).

[3] X. Z. Yu, N. Kanazawa, Y. Onose, K. Kimoto, W. Z. Zhang, S. Ishiwata, Y. Matsui, and Y. Tokura, Nat. Mater. **10**, 106 (2010).

[4] X. Z. Yu, N. Kanazawa, W. Z. Zhang, T. Nagai, T. Hara, K. Kimoto, Y. Matsui, Y. Onose, and Y. Tokura, Nat. Commun. **3**, 988 (2012).

[5] M. van Schilfgaarde, I. A. Abrikosov, and B. Johansson, Nature (London) **400**, 46 (1999).

[6] I. Raicevic, Dragana Popovic, C. Panagopoulos, L. Benfatto, M. B. Silva Neto, E. S. Choi, and T. Sasagawa, Phys. Rev. Lett. **106**, 227206 (2011).

[7] A. I. Liechtenstein, M. I. Katsnelson, V. P. Antropov, and V. A. Gubanov, J. Magn. Magn. Mater. **67**, 65 (1987).

[8] M. I. Katsnelson and A. I. Liechtenstein, J. Phys. Condens. Matter **16**, 7439 (2004).

[9] A. R. Machintosh and O. K. Andersen, in *Electrons at the Fermi Surface*, edited by M. Springford (Cambridge University Press, London, 1980), p. 149.

[10] M. Methfessel and J. Kubler, J. Phys. F **12**, 141 (1982).

[11] B. L. Györffy and G. M. Stocks, in *Electrons in Disordered Metals and Metallic Surfaces*, edited by P. Phariseau, B. L. Györffy, and L. Scheire (Plenum, New York, 1979).

[12] J. H. Mentink, J. Hellsvik, D. V. Afanasiev, B. A. Ivanov, A. Kirilyuk, A. V. Kimel, O. Eriksson, M. I. Katsnelson, and T. Rasing, Phys. Rev. Lett. **108**, 057202 (2012).

[13] Raghuveer Chimata, Anders Bergman, Lars Bergqvist, Biplab Sanyal, and Olle Eriksson, Phys. Rev. Lett. **109**, 157201 (2012).

[14] P. Lloyd, Proc. Phys. Soc. London, At. Mol. Phys. **90**, 207 (1967).

[15] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.111.127204 for calculation of the total energy variation in case of one- and two-site spin rotation in terms of a multiple scattering formalism and mapping the results onto effective spin Hamiltonians.

[16] S. Lounis and P. H. Dederichs, Phys. Rev. B **82**, 180404(R) (2010).

[17] A. Bergman, Ph.D. thesis, Uppsala University, 2006.

[18] O. Gunnarsson, O. Jepsen, and O. K. Andersen, Phys. Rev. B **27**, 7144 (1983).

[19] S. Frota-Pessoa, R. B. Muniz, and J. Kudrnovsky, Phys. Rev. B **62**, 5293 (2000).

[20] R. Pauthenet, J. Appl. Phys. **53**, 2029 (1982); **53**, 8187 (1982).

[21] G. Shirane, V. J. Minkiewicz, and R. Nathans, J. Appl. Phys. **39**, 383 (1968).

[22] J. W. Lynn, Phys. Rev. B **11**, 2624 (1975).

[23] R. Singer, F. Dietermann, and M. Fähnle, Phys. Rev. Lett. **107**, 017204 (2011).