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Letter to the Editor

On the energetics of transversal and longitudinal fluctuations of atomic magnetic moments

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ABSTRACT

By constrained spin-density functional calculations we estimate the relative role of the longitudinal and transversal fluctuations of the magnetic moments in the series of 3d metals (bcc Fe, hcp and fcc Co, and fcc Ni) for weak excitations from the ferromagnetic ground state. It is shown that the importance of longitudinal fluctuations strongly varies from relatively small in bcc Fe to large in fcc Ni. This means that a consistent adiabatic treatment of the low-energy spin fluctuations should include independent longitudinal fluctuations.

The magnetic interactions in solids can be accurately calculated by the ab-initio spin-density functional electron theory. Because this theory is very time-consuming and costly, the studies of dynamics and thermodynamics of itinerant-electron systems are usually based on parametrizations of the magnetic energy on the atomic level. Thereby the magnetic systems are subdivided into appropriately defined atomic volumes around the atoms. For time scales much larger than the interatomic electron hopping time $h/W \approx 10^{-15}$ s (h is Planck's quantum and W is the width of the electronic energy band in the solid) it is assumed (see, e.g., [1]) that one can average over the fast magnetic degrees of freedom in the atomic volumes and define atomic magnetic moments $\mathbf{M}_i = \mathbf{e}_i|\mathbf{M}_i|$ at the atomic sites i.

The ground state of the system is characterized by the ground-state magnetic moments $\{\mathbf{M}_i^g\}$ at all sites i. In a ferromagnetic ground state all these moments are parallel, in an antiferromagnetic ground state the orientations of the moments are antiparallel between various antiferromagnetic sublattices, and in more complicated systems (e.g., fcc Fe) there are noncollinear ground state orientations. In physical situations considering the spin dynamics or the thermodynamics at non-zero temperatures excitations of the $\{\mathbf{M}_i\}$ are involved. Examples are spin-wave configurations or disordered-moment systems far above the Curie temperature T_C. In principle these excitations may include transversal fluctuations of the moments, i.e., fluctuations of the $\{\mathbf{e}_i\}$, or longitudinal fluctuations, i.e., fluctuations of the $\{|\mathbf{M}_i|\}$.

In the so-called adiabatic approximation [2,3] it is assumed that the dynamics of the orientations of the directions of the atomic magnetic moments is so slow that the electronic system is at any instant in its ground state with respect to the orientations $\{\mathbf{e}_i\}$. Then the magnitudes $\{|\mathbf{M}_i|\}$ are "slaved" by the $\{\mathbf{e}_i\}$, i.e., there are only independent transversal fluctuations but no independent longitudinal fluctuations. To describe the interatomic exchange interactions, the simplest and most popular parametrization of the adiabatic exchange-energy is the classical Heisenberg model:

$$E(\{\mathbf{e}_i\}) = \sum_{i,j} J_{ij}\mathbf{e}_i \cdot \mathbf{e}_j, \tag{1}$$

where the J_{ij} are the exchange-interactions between all atomic pairs (i,j) in the system. Extensions of the Heisenberg model have been suggested (see, e.g., Ref. [4]), and a complete parametrization of the adiabatic exchange energy for systems without spin–orbit coupling and no external field has been given in [4] in terms of products of the two-spin basis functions $\mathbf{e}_i \cdot \mathbf{e}_j$. A complete representation of the total adiabatic magnetic energy (i.e., not just of the interatomic exchange energy) in terms of one-spin basis functions $\Phi_\nu(\{\mathbf{e}_i\})$ was given by the spin-cluster expansion [5] which was applied [6] to describe spin interactions in bcc and fcc Fe beyond the Heisenberg model.

The neglect of the longitudinal fluctuations is often justified by the argument that the intraatomic exchange (which "builds" the moments) is large compared to the interatomic exchange (which is responsible for the relative orientations of the moments), see, e.g., Ref. [7]. Unfortunately, it is not known how to calculate by the ab-initio density functional theory just the intraatomic exchange in a solid, because a modification of the $|\mathbf{M}_i|$ while fixing the $\mathbf{e}_i$ modifies both the intraatomic and the interatomic

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exchange. In the literature it therefore was attempted to investigate directly the relative importance of the longitudinal and transversal fluctuations instead of looking at the exchange interactions. The first calculations in this sense were performed by Uhl and Kübler [8] who used a Hamiltonian which describes magnetization fluctuations by a Ginzburg–Landau type expansion in terms of even powers of $\mathbf{M}(\mathbf{r})$ and an exchange-coupling term between the sites $\mathbf{r}$ and $\mathbf{r}'$ (instead of the gradient term occurring in the Ginzburg–Landau expansion). By using a Fourier-expansion of $\mathbf{M}(\mathbf{r})$, i.e., a representation of $\mathbf{M}(\mathbf{r})$ by frozen magnons they expressed $\hat{H}$ in Fourier space and determined all the parameters occurring in $\hat{H}$ by constrained spin-density functional calculations for frozen magnons. Then they calculated for Fe, Co and Ni the Curie temperature, using a mean-field approximation for the partition function calculated by means of this $\hat{H}$. The results demonstrated that both the longitudinal and the transversal fluctuations are important at the phase transition. Later, Ruban et al. [9] also demonstrated that both types of fluctuations are important in Fe and Ni at the phase transition. They used a real-space Hamiltonian for the two types of fluctuations similar to the one introduced by Shallcross et al. [10]. They calculated the parameters in this model also by the spin-density functional theory, considering not the ferromagnetic state as reference state but the disordered local moment state, and then investigated the phase transition by Monte Carlo simulations. Sandratskii [11] formulated for half-metallic NiMnSb a thermodynamical model which takes into account both fluctuation modes for temperatures up to about half of the Curie temperature, and he determined the energies of the fluctuations by the constrained spin-density functional theory. It was shown that the contributions of the two types of fluctuations to the temperature dependence of the Ni net magnetization compensate. In the present letter we give some more information on the energetics of longitudinal and transversal spin fluctuations which is obtained by the ab-initio spin-density functional electron theory. The data supplement the study of Ref. [9] in two respects. First, we consider not only bcc Fe and fcc Ni but also hcp Co and fcc Co. Second, we take as reference state not the randomly disordered local moment state which may be considered as appropriate effective medium close to and above T_C, but the ferromagnetic state.

To figure out the general importance of longitudinal and transversal fluctuations for the thermodynamics one would have to calculate the partition function [11]:

$$Z = \prod_i \int_0^{2\pi} \mathrm{d}\varphi_i \int_0^{\pi} \sin(\vartheta_i)\,\mathrm{d}\vartheta_i \int_0^{\infty} \mathrm{d}|\mathbf{M}_i|\,|\mathbf{M}_i|^2/\mu_B^3 \exp(-E(\{|\mathbf{M}_i|,\varphi_i,\vartheta_i\})/k_BT), \quad (2)$$

where E is the energy of a configuration $\{|\mathbf{M}_i|,\mathbf{e}_i\}$ given by the the atomic magnetic moments at all sites i, with the orientations $\{\mathbf{e}_i\}$ parametrized by the angles $\{\varphi_i,\vartheta_i\}$. For a ferromagnetic ground state we would have $|\mathbf{M}_i| = |\mathbf{M}_i^g|$ and $\vartheta_i = 0$ for all i. Longitudinal or transversal fluctuations of $\mathbf{M}_i$ would correspond to $|\mathbf{M}_i| \neq |\mathbf{M}_i^g|$ or $\vartheta_i \neq 0$ respectively. Eq. (2) shows that the importance of the two types of fluctuations is determined both by the energetics of these fluctuations and by the Jacobian weights $\sin(\vartheta_i)$ and $|\mathbf{M}_i|^2$. To determine Z one would have to calculate the energies $E(\{|\mathbf{M}_i|,\varphi_i,\vartheta_i\})$ for all conceivable configurations $\{|\mathbf{M}_i|,\varphi_i,\vartheta_i\}$. We confine ourselves to calculate ab-initio the energetics for the above discussed systems with ferromagentic ground state and small deviations of respectively one single atomic moment $\mathbf{M}_i$ of a supercell from the ground state values of $|\mathbf{M}_i|$ and ϑ_i, see below. We therefore cannot calculate the general partition function (2). However, we think that from the energetics of these single-atom longitudinal and transversal fluctuations we can estimate the relative importance of these fluctuations for weak exciations, e.g., for spin fluctuations at low temperatures. All calculations were performed by the linear-muffin-tin-orbital method [12] in atomic-sphere approximation and with the local-spin-density approximation part of the generalized gradient approximation of Ref. [13]. All calculations were in scalar relativistic approximation. The orientations $\mathbf{e}_i$ and the magnitudes $|\mathbf{M}_i|$ of the atomic moments $\mathbf{M}_i$ can be constrained by transversal and longitudinal Lagrangian fields [14]. For changes of the $\mathbf{e}_i$ the $|\mathbf{M}_i|$ were not constrained to the ground state moment $|\mathbf{M}_i^g|$ but instead calculated self-consistently (as in the adiabatic approximation). For fixed $\mathbf{e}_i$ the $|\mathbf{M}_i|$ are constrained by the longitudinal Lagrangian fields. The modifications of $\mathbf{e}_i$ or $|\mathbf{M}_i|$ are performed for just one atom of a supercell whereas the magnetic moments of the other atoms correspond to the ground state moments $\mathbf{M}_i^g$. For bcc Fe we used a two-atom cubic supercell consisting of nearest-neighbour atoms. For fcc systems we used a cubic four-atom supercell where the basis atoms are the one at the origin and the three neighbouring face-centering atoms. For hcp Co the usual two-atom supercell was used. For $|\mathbf{M}_i|$ we considered changes of the sizes $\delta = p\cdot 0.001|\mathbf{M}_i^g|/\mu_B$ with $p = 1,2,\ldots,10$. For the orientation the angle relative to the ferromagnetic alignment was changed by $\delta = p\cdot 0.001\pi$. For ferromagnets and small deviations of $\mathbf{M}_i$ from $\mathbf{M}_i^g$ the response of the energy on these deviations is expected to be proportional to δ^2, i.e., $\Delta E_{\mathrm{long}} = a_{\mathrm{long}}\delta^2$ for longitudinal and $\Delta E_{\mathrm{trans}} = a_{\mathrm{trans}}\delta^2$ for transversal fluctuations. The proportionality factors a_{long} and a_{trans} were obtained by quadratic fits of these expressions to the ab-initio calculated values of ΔE_{long} and $\Delta E_{\mathrm{trans}}$.

Table 1 shows the values of $|\mathbf{M}_i^g|$, a_{long} and a_{trans}. Our data demonstrate that the changes of the energy due to small longitudinal or transversal fluctuations are very similar. For hcp Co and especially for bcc Fe longitudinal fluctuations cost more energy than transversal fluctuations. This means that especially for bcc Fe the adiabatic approximation is good at least for weakly excited moment configurations. In contrast, for fcc Co and fcc Ni the longitudinal fluctuations should not be neglected.

To conclude, our ab-initio data for small deviations of the atomic magnetic moments $\mathbf{M}_i = \mathbf{e}_i\cdot|\mathbf{M}_i|$ from the ferromagnetically aligned ground-state moments $\mathbf{M}_i^g$ show that the related excitation energies are not very much different for longitudinal and transversal fluctuations in bcc Fe, hcp and fcc Co and fcc Ni. This is in contrast to the wide-spread assumption that the longitudinal fluctuations can be neglected because the intraatomic exchange interactions are considered to be large compared to the interatomic exchange interactions. Our results support the claim of Ref. [9] that both types of fluctuations are important for thermal excitations of the system of magnetic moments. This means that a consistent adiabatic treatment of low-energy spin fluctuations should include independent longitudinal fluctuations. Of course the conventional adiabatic approximation is always valid for the discussion of spin interactions in the ground state. At the end we mention what kind of physical effects may receive a better understanding by the above findings. In principle, all thermodynamic magnetic quantities are affected. In this sense

Table 1
The ground-state magnetic moments $|\mathbf{M}_i^g|$ and the proportionality factors a_{long} and a_{trans}, in units of mRy, for the excitation energies $\Delta E_{\mathrm{long,trans}} = a_{\mathrm{long,trans}}\delta^2$ for small deviations δ (see text) from the ferromagnetic ground state.

Material	$\lvert\mathbf{M}_i^g\rvert[\mu_B]$	a_{long}	a_{trans}
bcc Fe	2.26	139	43.8
hcp Co	1.64	57.0	51.3
fcc Co	1.67	29.6	44.8
fcc Ni	0.65	3.55	8.69

our results show that improved models of magnetism in itinerant magnets have to take into account independent longitudinal fluctuations. The relevant thermodynamic magnetic potential may be represented in terms of the N-point correlation functions for the adiabatic magnetic moments, and there are contributions to these correlation functions from longitudinal fluctuations. We are convinced that the agreement between experimental results for the correlation functions obtained, e.g., by magnetic neutron scattering or by x-ray absorption spectroscopy [15], and theoretical results obtained, e.g., by Monte Carlo simulations is improved when taking into account in the simulations the independent longitudinal fluctuations. In the present paper we discussed the important role of these fluctuations for weak thermal excitations, but we are convinced that they are similarly or even more important for strong thermal excitations. Therefore results for the critical temperature should become more reliable when including independent longitudinal fluctuations in the Monte Carlo simulations.

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