Adiabatic Dynamics of Local Spin Moments in Itinerant Magnets

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Using the adiabatic approximation, we derive the equations of motion for local spin moments which are valid for itinerant as well as tight-binding spins. Material parameters in the equations of motion can be obtained using standard density functional methods, because they depend only on the energy and Berry phase of the constrained ground state of frozen spin configurations. For the calculation of spin waves in a collinear magnet, it is sufficient to know the quadratic forms of total energy and spin component along the symmetry axis as functions of the spin deviations from the ground state configuration.

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Historically, there have been two opposing points of view of magnetism [1]: The atomic moment picture of Heisenberg and the itinerant electron picture of Bloch and Stoner. Because of its conceptual simplicity, the Heisenberg model has enjoyed a dominant position in the theory of magnetism, despite the tremendous success of the itinerant picture for transition metals. There have also been steady and serious efforts trying to push the atomic moment picture beyond its obvious domain of validity [2,3]. Recently, it has been proposed to use such local moments to formulate large amplitude spin dynamics which are needed for finite temperature effects on magnetism [4].

In this Letter, we show that while it is still possible to define local moments for itinerant spins, the usual Landau-Lifshitz equations for the moments must be replaced by more accurate equations of motion. This we establish purely based on the adiabatic assumption of the local moments with respect to the system's other degrees of freedom. For collinear magnets, the Berry curvatures involved in the equations of motion are shown to describe how the total spin component along the symmetry axis changes due to spin deviations from the ground state configuration. A simple formula for the spin wave spectrum, previously derived for ferromagnets in either the free or tight-binding limits, is now shown to be valid everywhere between these limits and for all collinear magnets.

Adiabatic dynamics of the local moments.—Following the usual practice, we partition the space into spin cells of volume V_j [5]. There is some empirical knowledge as to how to choose the partition effectively, but one does not have to assign one cell for each atom. We concentrate first on how to derive the best set of equations of motion in the adiabatic limit for a given partition. The spin for a cell is defined as $\mathbf{s}_j = \langle\psi|\hat{\mathbf{s}}_j|\psi\rangle$, where $\hat{\mathbf{s}}_j = \int_{V_j} d^3r\, \hat{\mathbf{s}}(\mathbf{r})$, and $\hat{\mathbf{s}}(\mathbf{r})$ is the spin density operator. (In this Letter, we use the convention that spin angular momenta have an extra factor of $\hbar$ beside s.) Let $|\psi[\mathbf{s}]\rangle$ be the lowest energy state of the electronic system which yields a given spin configuration denoted by $[\mathbf{s}]$. Using a similar method as in [6], we find

$$-\sum_{\alpha' j'} \hbar\Omega^{\alpha\alpha'}_{jj'}\dot{s}^{\alpha'}_{j'} + \frac{\partial E}{\partial s^{\alpha}_{j}} = 0\,, \tag{1}$$

where α labels the spin components, E is the energy of the constrained ground state, and the Ω matrix elements (Berry curvatures [7]) are defined as

$$\Omega^{\alpha\alpha'}_{jj'} = \frac{\partial}{\partial s^{\alpha}_{j}}\left\langle\psi\left|\frac{i\partial}{\partial s^{\alpha'}_{j'}}\right|\psi\right\rangle - \frac{\partial}{\partial s^{\alpha'}_{j'}}\left\langle\psi\left|\frac{i\partial}{\partial s^{\alpha}_{j}}\right|\psi\right\rangle. \tag{2}$$

Therefore, the adiabatic dynamics of the spin moments is completely determined from the energy and the Berry curvatures of the constrained ground state. These quantities can be extracted using standard density functional methods [8], which are simpler and thus more practical than methods based on the linear response or time-dependent density functional theory [9]. There has already been much work on the calculation of the energy [5], and one also has a few tricks to extract the Berry curvatures [6]. For spin wave calculations, the Berry curvatures are needed only at the ground state, and we will show in this Letter that there is a powerful method to extract them when the spin ground state has an axial symmetry.

The equations of motion (1) are exact in the adiabatic limit [10] for a given partition of the space into cells, and are valid beyond the harmonic approximation or linear response. Based on the variational nature of the theory, the local moment description of the spins becomes better for finer partition of the space, simply because more degrees of freedom are included in the variational wave function [11]. There is nothing intrinsically wrong with the local moment description of itinerant spins; the specification of local moments for every point of space becomes equivalent to the description of spin configuration in terms of its Fourier components as used in [6].

The Landau-Lifshitz equations.—In the special situation of tight-binding or rigid spins, the wave function

$|\psi[\mathbf{s}_j]\rangle$ is obtained from the absolute ground state of spin configuration $\mathbf{s}_j^0$ by rigidly rotating the spins within each cell, i.e.,

$$|\psi[\mathbf{s}_j]\rangle = \prod_j e^{i\vec{\theta}_j\cdot\hat{\mathbf{s}}_j}|\psi_0\rangle, \tag{3}$$

where $\vec{\theta}_j$ is in the direction of $\mathbf{s}_j^0 \times \mathbf{s}_j$ and has a magnitude given by the angle between $\mathbf{s}_j$ and $\mathbf{s}_j^0$. The spin operators $\hat{\mathbf{s}}_j$ are for the total spin in each cell, and they satisfy the usual commutation relations for angular momenta, i.e., $[\hat{s}_j^\alpha, \hat{s}_{j'}^{\alpha'}] = i\delta_{jj'}\epsilon_{\alpha\alpha'\beta}\hat{s}_j^\beta$, where $\epsilon_{\alpha\beta\gamma}$ is the fundamental antisymmetric tensor. Then, the Berry curvatures can be calculated as

$$\Omega_{jj'}^{\alpha\alpha'} = \delta_{jj'}\epsilon^{\alpha\alpha'\beta}s_j^\beta/(\mathbf{s}_j)^2, \tag{4}$$

which says that there is no Berry curvature between different cells, and that the Berry curvature within a single cell is given by that of a rigid spin [12]. With this form of the Berry curvature, our general equations of motion (1) reduce to the classical Landau Lifshitz equations

$$\hbar\dot{\mathbf{s}}_j = \mathbf{s}_j \times \frac{\partial E}{\partial \mathbf{s}_j}. \tag{5}$$

However, the wave function (3) is not the constrained ground state for the given spin configuration in general. The Landau-Lifshitz equations can thus fail for itinerant spins, or when the local moments interact with itinerant electrons [13]. There are at least two ways that intermoment Berry curvatures arise and thus invalidate the Landau-Lifshitz equations. (i) In the presence of electron hopping, the moments at different sites may share the same spin and thus have Berry curvatures between them. (ii) Additional Berry phases may arise due to the adiabatic following of the neglected degrees of freedom to the local moments.

To illustrate point (i), we have solved exactly a $t - J - B$ Hamiltonian on two sites,

$$-\frac{t}{2}\sum_\sigma [a_{1\sigma}^\dagger a_{2\sigma} + a_{2\sigma}^\dagger a_{1\sigma}] - 4J\hat{\mathbf{s}}_1\cdot\hat{\mathbf{s}}_2 - \sum_j \mathbf{B}_j\cdot\hat{\mathbf{s}}_j,$$

where $a_{j\sigma}$ is the destruction operator on site j and spin state σ. When there are two electrons present in the system, there are six different states, four with one electron on each site and two with both electrons on either site. A uniform magnetic field, $\mathbf{B}_j = B\hat{z}$, is added to select a direction of magnetization for the ferromagnetic ground state, and our final results have a well defined limit as $B \to 0$. There are six eigenstates consisting of a triplet with energies $-J - B, -J$, and $-J + B$, and of three singlets with energies at 0 and $\frac{3}{2}J \pm [(\frac{3}{2}J)^2 + (2t)^2]^{1/2}$. If the hopping energy is not too large ($t^2 < J^2 + 5BJ + B^2$), the ground state is one of the triplet, in which the average spin on each site is in the z direction with the full magnitude of $1/2$.

The average spins are tilted when small transverse fields are added to the system. The new ground state can be calculated perturbatively, from which the Berry curvatures at $\mathbf{B}_j = B\hat{z}$ are extracted exactly. The calculations are tedious but straightforward, and we have obtained the following simple results:

$$\Omega_{11}^{xy} = \Omega_{22}^{xy} = -2 - \frac{t^2}{(J+B)^2}, \tag{6}$$

$$\Omega_{12}^{xy} = \Omega_{21}^{xy} = \frac{t^2}{(J+B)^2}, \tag{7}$$

where the xx and yy components all vanish because of the rotational symmetry about the spin z axis. These results clearly demonstrate the effect of intersite hoppings. When t is zero, we go back to the original results of the tight-binding limit. When t is nonzero, not only the on-site Berry curvature is modified, but also an intersite Berry curvature is induced.

To illustrate point (ii), we consider the three spin model $H = -J(\mathbf{s}_1\cdot\mathbf{s}_2 + \mathbf{s}_2\cdot\mathbf{s}_3) - B(s_1^z + s_2^z + s_3^z)$. Two of these spins (1 and 3) will be taken into account in the definition of the local moments, and the other simulates a neglected degree of freedom such as an orbital moment or an interstitial spin pocket. The eigenstates consist of a quartet with energies $-2J \pm 2B$ and $-2J \pm B$, and of two doublets with energies $\pm B$ and $4J \pm B$, respectively. For ferromagnetic coupling ($J > 0$), the ground state is one of the quartet, with all the spins fully in the z direction ($B > 0$). We apply small transverse fields on spins 1 and 3 (leaving spin 2 free), causing transverse deviations of the spins in the new ground state. The Berry curvatures with respect to the transverse components of spins 1 and 3 are then calculated, with the results

$$\Omega_{11}^{xy} = \Omega_{33}^{xy} = -2\frac{B^2 + 4BJ + 5J^2}{(B+2J)^2}, \tag{8}$$

$$\Omega_{13}^{xy} = \Omega_{31}^{xy} = -2\frac{J^2}{(B+2J)^2}. \tag{9}$$

Significance of the Berry curvatures.—A transparent physical picture for the Berry curvatures can be obtained if we confine our attention to the case of ground states with rotational symmetry of the spins about an axis. This includes the usual ferromagnets, ferrimagnets, and antiferromagnets, but excludes canted or spiral magnetic structures. No translational symmetry will be assumed except in a special discussion. The Berry curvatures will be related to the reduction of the total spin z component due to the excitations of the spins, and a simple formula for the frequency dispersion will be obtained.

Let the symmetry axis of the ground state be the z direction, then the xy spin components due to a spin wave eigenmode can be written as [14]

$$\mathbf{s}_j^\perp = \mathrm{Re}[e^{-i\omega t}(\hat{x} + i\hat{y})s_j], \tag{10}$$

where s_j is the amplitude (complex) on the jth site. Using the complex amplitudes in Eq. (10) as the independent variables, we can obtain the same set of equations of

motion as in Eq. (1), except that the upper indices now refer to the real and imaginary parts of the amplitudes. By linearizing the energy gradient and writing $K_{jj'}^{\alpha\alpha'}$ for the second derivative of the energy with respect to s_j^{α} and $s_{j'}^{\alpha'}$, we obtain the eigenmode equations as

$$\sum_{j'} [i\hbar\omega\Omega_{jj'} + K_{jj'}]s_{j'} = 0, \tag{11}$$

where $\Omega_{jj'} = \Omega_{jj'}^{11} - i\Omega_{jj'}^{12}$, and $K_{jj'} = K_{jj'}^{11} - iK_{jj'}^{12}$.

Therefore, once we know the K and Ω matrices, the above equations of motion determine the spin wave spectrum and the associated eigenmodes. It is easy to see that the quadratic form $\frac{1}{2}s^{\dagger}Ks = \frac{1}{2}\sum_{jj'} s_j^* K_{jj'}s_{j'}$ represents the energy ΔE for creating the spin excitations $\{s_j\}$. The diagonal element K_{jj} can thus be extracted from the energy increase in the system when only a single spin s_j is excited. The off-diagonal elements $K_{jj'}$ and $K_{j'j} = K_{jj'}^*$ $(j \neq j')$ can be obtained from the energy increase when two spins are excited. How about the Ω matrix? In the following, we will show that $-i\frac{1}{2}s^{\dagger}\Omega s = -i\frac{1}{2}\sum_{jj'} s_j^*\Omega_{jj'}s_{j'}$ is, in fact, the quadratic form for the reduction, $\Delta S_z = S_z^0 - \langle\hat{S}_z\rangle$, of the total spin z component of the ground state due to the spin excitations, i.e.,

$$-i\frac{1}{2}s^{\dagger}\Omega s = \Delta S_z. \tag{12}$$

This relation then allows a simple evaluation of the Ω matrix in a similar manner as for the K matrix, and it is not necessary to invoke directly the constrained ground state wave functions, which are usually much harder to obtain than the spin expectation values.

That the relation (12) is valid for all collinear magnets can be easily seen in the tight-binding limit. The spin configuration of a collinear magnetic ground state may be written as $\mathbf{s}_j = s_j^0\hat{z}$, where s_j^0 can be positive (ferromagnet) or take both signs (ferrimagnets or antiferromagnets). The total reduction of the spin-z component due to a spin wave (10) is just $\Delta S_z = \frac{1}{2}\sum_j |s_j|^2/s_j^0$. The same result is obtained from the left-hand side of Eq. (12), if we use $\Omega_{jj'} = -i\Omega_{jj'}^{12} = i\Omega_{jj'}^{xy} = i\delta_{jj'}(s_j^0)^{-1}$, which is justified according to Eq. (4) and from the rotational invariance of the ground state about the z axis.

To establish the relation (12) beyond the tight-binding limit, we first show that $i\frac{1}{2}s^{\dagger}\Omega s$ is actually a Berry phase in the complex plane of the overall amplitude of the spin wave. Let us write $s_j = AR_j$, where $A = A_1 + iA_2$ is the overall amplitude, and $R_j = R_j^1 + iR_j^2$ are the relative amplitudes which are regarded as known and fixed. The Berry curvature at the origin of the complex A plane is given by

$$\Omega_A = \frac{\partial}{\partial A_1}\left\langle\psi\left|\frac{i\partial}{\partial A_2}\right|\psi\right\rangle - \frac{\partial}{\partial A_2}\left\langle\psi\left|\frac{i\partial}{\partial A_1}\right|\psi\right\rangle. \tag{13}$$

The derivatives can be transferred to those with respect to the s_j variables, with the result

$$\sum_{jj'}[\Omega_{jj'}^{11}(R_j^2R_{j'}^1 - R_j^1R_{j'}^2) + \Omega_{jj'}^{12}(R_j^1R_{j'}^1 + R_j^2R_{j'}^2)],$$

where we have used the relations $\Omega_{jj'}^{11} = \Omega_{jj'}^{22}$ and $\Omega_{jj'}^{12} = -\Omega_{jj'}^{21}$, due to the rotational symmetry of the ground state. On the other hand, by expanding in terms of the real and imaginary parts of A and R_j, we find that $i\frac{1}{2}s^{\dagger}\Omega s$ is just $\frac{1}{2}|A|^2$ times the expression (13), or the Berry phase around a small loop of area $\frac{1}{2}|A|^2$.

Next we show that this Berry phase is, in fact, equal to the reduction of the total spin-z component. Consider a circular wedge of radius $|A|$ and angle ϕ. The Berry phase around the sides of this wedge is just Ω_A (13) times the area $\frac{1}{2}\phi|A|^2$. It can also be calculated directly as the phase of $\langle\psi|\psi_\phi\rangle$, where $|\psi\rangle$ is the constrained ground state for the case of $A_1 = |A|, A_2 = 0$, while $|\psi_\phi\rangle$ is for $A_1 = |A|\cos\phi, A_2 = |A|\sin\phi$. Here we have taken the phase convention of wave functions such that their overlaps with the ground state are real and positive. Because a phase rotation of the amplitude A is equivalent to a rotation of the total spin about the z axis, we may write $|\psi_\phi\rangle = e^{i\phi(\hat{S}_z - S_z^0)}|\psi\rangle$, where, in order to be consistent with the above phase convention, we have subtracted from $\hat{S}_z$ its expectation value S_z^0 in the ground state. For sufficiently small ϕ, we may expand the exponential of the spin rotation operator, and find that the Berry phase along the wedge is equal to $\phi(\langle S_z\rangle - S_z^0)$ as desired.

A simple formula for the spinwave spectrum.—Multiplying s_j^* on the terms of Eq. (11), and summing over j, we find that the eigenfrequency can be expressed as

$$\hbar\omega = \frac{\frac{1}{2}s^{\dagger}Ks}{-i\frac{1}{2}s^{\dagger}\Omega s} = \frac{\Delta E}{\Delta S_z}, \tag{14}$$

where we have used the relations of the quadratic forms of the K and Ω matrices to the energy increase and spin-z reduction of the ground state due to the spin excitations [15]. This was obtained by Niu and Kleinman [6] for the special cases of ferromagnets in the jellium and tight-binding limits. It is now shown to be valid everywhere between those limits, and to be valid for all collinear magnets including ferrimagnets and antiferromagnets. Because our derivation does not depend on how the space is partitioned into cells, this formula is an exact result of the adiabatic limit and the axial symmetry of the ground state.

There is, in fact, a deep connection between the above simple formula and the quantum theory of spin waves. According to the latter, the energy of a magnon (the spin wave quantum) is just $\hbar\omega$, and there is a single spin flip from the ground state per magnon in a ferromagnet. The above formula simply says that the magnon energy is given by the energy of the spin wave divided by the number of magnons in the spin wave. Interestingly, the connection also applies to ferrimagnetic and antiferromagnetic spin waves. In the latter case, the ground state magnetization S_0^z is zero, but $\langle\hat{S}_z\rangle$ is

finite in a spin wave of the form (10) [14]. This can be verified directly from the classical spin wave solutions of the Heisenberg model [16]. The fact that the spectrum goes linearly with k at small k is explained by the observation that $\langle \hat{S}_z \rangle$, which enters the denominator of the formula (14), vanishes linearly as $k \to 0$. Quantum mechanically [16], the z component of the total spin, which is a good quantum number, has eigenvalues of ± 1 in each degenerate subspace of one-magnon states, where the negative eigenvalue corresponds to a spin wave of the form (10), while the positive eigenvalue corresponds to a spin wave of the opposite chirality.

The formula (14) can also be used to evaluate the spin wave spectrum. Suppose, for instance, the cell spins in the ground state form a Bravais lattice; then one knows the spin wave eigenmode for each wave vector purely based on the translational symmetry of the system. All one needs to do is to calculate the energy increase due to the presence of the spin wave and the corresponding reduction of the total spin z component, and take the ratio of the two. This method has been used implicitly by Halilov *et al.* [5] and explicitly by Brown *et al.* [18] for the evaluation of spin wave spectra of the ferromagnets of crystalline Ni, Co, and Fe.

The only approximation in this method is the assumption of adiabaticity regarding the motion of the cell spins relative to the rest of the degrees of freedom in the system. One type of such degrees of freedom are spin fluctuations within each cell, and they become important when there are more than one atom per cell. In such a case, one should take a finer partition of space for accuracy, and pay the price of not being able to know the spin wave eigenmodes based on symmetry arguments alone. One may still use the formula (14) to calculate the spin wave spectrum variationally based on trial spin wave configurations. Of course, one can always use Eq. (11) or even (1) to obtain the spectrum systematically.

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[10] This condition is satisfied at the long wavelength limit of a spin wave, but is violated in the region of the so-called Stoner continuum where the spin waves are heavily damped.

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