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Role of quadratic terms in the Heisenberg model for quantum spin dynamics

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The dynamics of the expectation value of a spin operator $\hat{\mathbf{S}}_n$ in the quantum mechanical Heisenberg model has been compared with the trajectory of the corresponding classical spin $\mathbf{S}_n$. A Hamilton operator $\hat{\mathcal{H}}$ linear in the spin operator $\hat{\mathbf{S}}_n$ shows a perfect agreement between quantum mechanical and classical trajectories. Quadratic or higher-order terms in the Hamilton operator lead to disagreement. The corresponding correction term can be found for different physical descriptions and methods dealing with the quantum mechanical Heisenberg model.

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I. INTRODUCTION

Micromagnetic simulations are a common tool to investigate, e.g., the dynamics of domain walls[1–3] or the reversal mechanism of a magnetic nanoparticle.[4,5] While magnetism is a pure quantum mechanical phenomenon that can not be described by classical physics, mostly all spin-dynamics simulations are dealing with a classical description. The explanation for this is manifold. At the beginning, micromagnetic simulations were introduced as an approximation caused by the fact that the correct quantum mechanical description is impossible or too complex.[6,7] The approximation was strengthened by experimental results showing a good agreement with the micromagnetic predictions.

As a reason for the agreement, we can mention, on the one hand, the correspondence principle,[8] which states that if the quantum numbers describing the system are large, an agreement between quantum and classical physics occurs. On the other hand, we have the Ehrenfest theorem,[9] which states that the expectation values obey the classical dynamical laws. However, a satisfying answer to the question, in which case it is possible to use a classical equation such as the Landau-Lifshitz-Gilbert[10–12] (LLG) or the Bloch equation[13] to describe the spin dynamics, has not been given so far. To answer this question, we have investigated the dynamics of quantum mechanical spins and compared the trajectories of their expectation values with the trajectories of classical spins using the according classical Heisenberg Hamiltonian.

The paper is organized as follows. In Sec. II, we give a simple rule that allows us to decide if the dynamics behaves classical or not, followed by a first analytical proof of our theorem. Due to the complexity of the underlying differential equations, the analytical consideration was restricted to linear excitations. A further proof for nonlinear situations will be given by numerical methods in Sec. III. After that, Sec. IV brings the previous results into a bigger (historical) context. Section V gives a short summary followed by an appendix, which describes an alternative way for calculating ferromagnetic spin waves in a quantum mechanical Heisenberg model with single-ion anisotropy solving the Schrödinger equation.

II. ANALYTICAL CONSIDERATION

The probably best starting point for analytical considerations of the dynamics of quantum spins is the Heisenberg equation of motion

$$i\hbar\frac{\partial\hat{\mathbf{S}}_n}{\partial t} = [\hat{\mathbf{S}}_n,\hat{\mathcal{H}}]. \tag{1}$$

It is known[7,14–16] that this differential equation for the Heisenberg Hamiltonian $\hat{\mathcal{H}}$ with an additional Zeeman term

$$\hat{\mathcal{H}} = -\frac{1}{2\hbar^2}\sum_{l,m} J_{lm}\hat{\mathbf{S}}_l\cdot\hat{\mathbf{S}}_m - \mu_S B_z\sum_l \hat{S}_l^z \tag{2}$$

can be written as

$$\frac{\partial\hat{\mathbf{S}}_n}{\partial t} = \hat{\mathbf{S}}_n\times\hat{\mathbf{H}}_n \tag{3}$$

with an effective field $\hat{\mathbf{H}}_n = -\mathrm{grad}_{\hat{\mathbf{S}}_n}\hat{\mathcal{H}}$. This equation looks like the famous Landau-Lifshitz equation[17] without damping. Therefore, it is the starting point of any classic consideration concerning spin dynamics. However, the terms are still operators and not classical numbers. Furthermore, all the terms of the Hamiltonian equation (2) are linear in $\hat{\mathbf{S}}_n$. This is an important point, as it means that $\hat{\mathbf{H}}_n$ does not contain $\hat{\mathbf{S}}_n$ anymore and, therefore, the commutator $[\hat{\mathbf{S}}_n,\hat{\mathbf{H}}_n]$ is zero. However, terms quadratic in $\hat{\mathbf{S}}_n$ such as single-ion anisotropies [e.g., $D_z(\hat{\mathbf{S}}_n)^2$] or the biquadratic exchange $J_{\mathrm{bi}}(\hat{\mathbf{S}}_n\cdot\hat{\mathbf{S}}_m)^2$ will lead to an effective field $\hat{\mathbf{H}}_n$, which depends on $\hat{\mathbf{S}}_n$. In this case, we have to deal with commutators such as $[\hat{S}_n^x,\hat{S}_n^y] = i\hbar\hat{S}_n^z$ (and cyclic permutations) and therefore $[\hat{\mathbf{S}}_n,\hat{\mathbf{H}}_n]\neq 0$.

The consequence is the occurrence of a correction term of the order of $\hbar$ in the resulting Heisenberg equation (3):

$$\frac{\partial\hat{\mathbf{S}}_n}{\partial t} = \hat{\mathbf{S}}_n\times\hat{\mathbf{H}}_n + \mathcal{O}(\hbar). \tag{4}$$

Alternatively, one can write Eq. (4) as

$$\frac{\partial\hat{\mathbf{S}}_n}{\partial t} = \frac{1}{2}(\hat{\mathbf{S}}_n\times\hat{\mathbf{H}}_n - \hat{\mathbf{H}}_n\times\hat{\mathbf{S}}_n). \tag{5}$$

This becomes more clear in the case of the following example: the Heisenberg equation for the Heisenberg Hamiltonian $\hat{\mathcal{H}}$ with additional single-ion anisotropy:

$$\begin{aligned}\hat{\mathcal{H}} &= -\frac{1}{2\hbar^2}\sum_{l,m} J_{lm}\hat{\mathbf{S}}_l\cdot\hat{\mathbf{S}}_m - D_z\sum_l\left(\hat{S}_l^z\right)^2\\ &= \hat{\mathcal{H}}_J + \hat{\mathcal{H}}_{D_z}.\end{aligned} \tag{6}$$

Here, we can write the Heisenberg equation as

$$i\hbar\frac{\partial\hat{\mathbf{S}}_n}{\partial t} = \{[\hat{\mathbf{S}}_n,\hat{\mathcal{H}}_J] + [\hat{\mathbf{S}}_n,\hat{\mathcal{H}}_{D_z}]\}. \tag{7}$$

The first term on the right-hand side can be directly written utilizing Eq. (3) as

$$[\hat{\mathbf{S}}_n,\hat{\mathcal{H}}_J] = i\hbar\sum_m\frac{J_{lm}}{\hbar^2}\hat{\mathbf{S}}_n\times\hat{\mathbf{S}}_m = -i\hbar\hat{\mathbf{S}}_n\times\hat{\mathbf{H}}_n^J, \tag{8}$$

again $\hat{\mathbf{H}}_n^J = -\mathrm{grad}_{\hat{\mathbf{S}}_n}\hat{\mathcal{H}}_J$. The second commutator contains the quadratic Hamiltonian of the single-ion anisotropy:

$$\begin{aligned}[\hat{\mathbf{S}}_n,\hat{\mathcal{H}}_{D_z}] &= -\frac{D_z}{\hbar^2}\sum_l\begin{pmatrix}[\hat{S}_n^x,\hat{S}_l^z\hat{S}_l^z]\\ [\hat{S}_n^y,\hat{S}_l^z\hat{S}_l^z]\\ [\hat{S}_n^z,\hat{S}_l^z\hat{S}_l^z]\end{pmatrix}\\ &= i\hbar\frac{D_z}{\hbar^2}\left\{\begin{pmatrix}\hat{S}_n^y\hat{S}_n^z\\ -\hat{S}_n^x\hat{S}_n^z\\ 0\end{pmatrix} + \begin{pmatrix}\hat{S}_n^z\hat{S}_n^y\\ -\hat{S}_n^z\hat{S}_n^x\\ 0\end{pmatrix}\right\}\\ &= \frac{i\hbar}{2}\left\{\hat{\mathbf{S}}_n\times\hat{\mathbf{H}}_n^{D_z} - \hat{\mathbf{H}}_n^{D_z}\times\hat{\mathbf{S}}_n\right\}\end{aligned} \tag{9}$$

with $\hat{\mathbf{H}}_n^{D_z} = -\mathrm{grad}_{\hat{\mathbf{S}}_n}\hat{\mathcal{H}}_{D_z}$. It is not difficult to see that this equation corresponds directly to Eq. (5). To get an equation similar to Eq. (3), one has to interchange $\hat{\mathbf{H}}_n^{D_z}$ with $\hat{\mathbf{S}}_n$ in the first term on the right-hand side of Eq. (9); therefore, we need the commutator $[\hat{\mathbf{S}}_n,\hat{\mathbf{H}}_n^{D_z}]$.

The interchange between $\hat{\mathbf{H}}_n^{D_z}$ and $\hat{\mathbf{S}}_n$ will be realized directly by the commutation of $\hat{S}_n^x$ respectively $\hat{S}_n^y$ with $\hat{S}_n^z$:

$$\begin{aligned}[\hat{\mathbf{S}}_n,\hat{\mathcal{H}}_{D_z}] &= i\hbar\frac{D_z}{\hbar^2}\left\{2\begin{pmatrix}\hat{S}_n^y\hat{S}_n^z\\ -\hat{S}_n^x\hat{S}_n^z\\ 0\end{pmatrix} + i\hbar\begin{pmatrix}\hat{S}_n^x\\ \hat{S}_n^y\\ 0\end{pmatrix}\right\}\\ &= i\hbar\left\{\hat{\mathbf{S}}_n\times\hat{\mathbf{H}}_n^{D_z} + i\frac{D_z}{\hbar}\begin{pmatrix}\hat{S}_n^x\\ \hat{S}_n^y\\ 0\end{pmatrix}\right\}.\end{aligned} \tag{10}$$

Connecting Eqs. (8), (10), and (7), we end up with a Heisenberg equation similar to Eq. (4):

$$\frac{\partial\hat{\mathbf{S}}_n}{\partial t} = \hat{\mathbf{S}}_n\times\left(\hat{\mathbf{H}}_n^J + \hat{\mathbf{H}}_n^{D_z}\right) + i\frac{D_z}{\hbar}\begin{pmatrix}\hat{S}_n^x\\ \hat{S}_n^y\\ 0\end{pmatrix} \tag{11}$$

with correction term $\mathcal{O}(\hbar) = iD_z/\hbar\,(\hat{S}_n^x\mathbf{x} + \hat{S}_n^y\mathbf{y})$.

The corresponding classical Landau-Lifshitz equation is given by

$$\frac{\partial\mathbf{S}_n}{\partial t} = \frac{\gamma}{\mu_S}\mathbf{S}_n\times\left(\mathbf{H}_n^J + \mathbf{H}_n^{D_z}\right) \tag{12}$$

with the effective fields $\mathbf{H}_n^\alpha = -\mathrm{grad}_{\mathbf{S}_n}\mathcal{H}_\alpha$, $\alpha\in\{J,D_z\}$, and the gyromagnetic ratio $\gamma = g\mu_B/\hbar$. It should be noticed that, in the classical description, the spins $\mathbf{S}_n = \boldsymbol{\mu}_n/\mu_S$, as well as the effective fields, are ordinary numbers (vectors) and not operators. Furthermore, the classical Hamilton functions $\mathcal{H}_\alpha$ look similar to the Hamilton operators $\hat{\mathcal{H}}_\alpha$ [Eqs. (2) and (6)].

To discuss the difference between classical and the quantum mechanical spin dynamics, we have to solve both equations (the classical Landau-Lifshitz and the quantum mechanical Heisenberg equation). Due to the minor differences between these two equations, we solve both at the same time. Therefore, we need some abbreviations. In the following, the S_n^α are either the x or y component ($\alpha\in\{x,y\}$) of the classical spin $\mathbf{S}_n$ or the corresponding quantum mechanical expectation values $\langle\hat{S}_n^\alpha\rangle$. Further, S is either the z component $S_n^z = S_z$ of the classical spin or the expectation value $\langle S_n^z\rangle\approx S$, with spin quantum number S of the single spin. Additionally, the variables η and ξ are given by $\eta = \gamma/\mu_S$ and $\xi = 0$ in the classical description (Landau-Lifshitz equation) and in the quantum mechanical picture (Heisenberg equation) η and ξ are given by $\eta = 1/\hbar$ and $\xi = 1$.

Using these abbreviations, the S_n^x and S_n^y contributions of the remaining differential equation can be written as

$$\frac{\partial\mathsf{S}_n^x}{\partial t} \approx \eta\left[\sum_m J_{nm}\mathsf{S}\left(\mathsf{S}_m^y - \mathsf{S}_n^y\right) + 2D_z\mathsf{S}\mathsf{S}_n^y + i\xi D_z\mathsf{S}_n^x\right], \tag{13a}$$

$$\frac{\partial\mathsf{S}_n^y}{\partial t} \approx -\eta\left[\sum_m J_{nm}\mathsf{S}\left(\mathsf{S}_m^x - \mathsf{S}_n^x\right) + 2D_z\mathsf{S}\mathsf{S}_n^x - i\xi D_z\mathsf{S}_n^y\right]. \tag{13b}$$

Then, we get (with $\mathsf{S}_n^\pm = \mathsf{S}_n^x \pm i\mathsf{S}_n^y$)

$$\begin{aligned}&\frac{\partial\mathsf{S}_n^\pm}{\partial t}\\ &\quad = \mp i\eta\left[\sum_m J_{nm}\mathsf{S}\left(\mathsf{S}_m^\pm - \mathsf{S}_n^\pm\right) + 2D_z\mathsf{S}\mathsf{S}_n^\pm - \xi D_z\mathsf{S}_n^\pm\right].\end{aligned} \tag{14}$$

And finally, with the Fourier transformations

$$\mathsf{S}_n^\pm = \sum_{\mathbf{k}}\mathsf{S}^\pm(\mathbf{k})e^{-i\mathbf{k}\cdot\mathbf{r}_n},$$

$$J(\mathbf{k}) = J(-\mathbf{k}) = \sum_m J_{nm}e^{i\mathbf{k}\cdot(\mathbf{r}_n-\mathbf{r}_m)},$$

the equation of motion becomes

$$\frac{\partial\mathsf{S}^\pm(\mathbf{k})}{\partial t} = \pm i\eta\left[\mathsf{S}[(J(\mathbf{0}) - J(\mathbf{k})] + 2D_z\left(\mathsf{S} - \frac{1}{2}\xi\right)\right]\mathsf{S}^\pm(\mathbf{k}). \tag{15}$$

If we further assume an exchange interaction between nearest neighbors only, we get

$$\frac{\partial\mathsf{S}^\pm(\mathbf{k})}{\partial t} = \pm i\omega\mathsf{S}^\pm(\mathbf{k}) \tag{16}$$

with

$$\omega = 2\eta\mathsf{S}J[1 - \cos(ka)] + 2D_z\left(\mathsf{S} - \frac{1}{2}\xi\right). \tag{17}$$

While the first term in Eq. (17) corresponds to the energy contribution coming from the exchange interaction, the second term describes the influence of the single-ion anisotropy and determines the resulting spin-wave gap. The $S - \frac{1}{2}$ in the quantum mechanical description ($\xi = 1$ and $\mathsf{S} = S$) underlines the well-known fact that systems with spin quantum number $S = \frac{1}{2}$ are not influenced by single-ion anisotropies.

The solutions of Eq. (16) are plane waves with wave vector $\mathbf{k}$ and energy dispersion $E(\mathbf{k}) = \hbar\omega(\mathbf{k})$:

$$\mathsf{S}^{\pm}(\mathbf{k}) = e^{\pm\frac{i}{\hbar}Et} = e^{\pm i\omega t}. \tag{18}$$

Then, the inverse Fourier transformation leads to

$$\begin{aligned}\mathsf{S}_n^{\pm} &= \mathsf{S}_n^x \pm i\mathsf{S}_n^y = e^{\pm i(\mathbf{k}\cdot\mathbf{r}_n - \omega t)}\\ &= \cos(\mathbf{k}\cdot\mathbf{r}_n - \omega t) \pm i\sin(\mathbf{k}\cdot\mathbf{r}_n - \omega t)\end{aligned}$$

and, finally, to

$$\begin{aligned}\mathsf{S}_n^x &\approx \mathsf{S}_\perp \cos(\mathbf{k}\cdot\mathbf{r}_n - \omega t),\\ \mathsf{S}_n^y &\approx \mathsf{S}_\perp \sin(\mathbf{k}\cdot\mathbf{r}_n - \omega t),\\ \mathsf{S}_n^z &\approx \mathsf{S}.\end{aligned} \tag{19}$$

The prefactor $\mathsf{S}_\perp$ appears due to the normalization.

III. NUMERICAL CONSIDERATION

Due to the complexity of the underlying differential equations, the previous analytical calculations have been restricted to linear excitations (spin waves). To prove the validity also in case of complex nonlinear excitations, we have to perform additional numerical calculations. To get a larger distance to the classical description, we switch from the Heisenberg to the Schrödinger picture. Therewith, we have to solve the time-dependent Schrödinger instead of the Heisenberg equation. This enables us to get an additional proof. Further, due to this change, we reduce the complexity of the numerical procedure. In the following, we give a detailed description of the numerical methods used.

In general, any spin state can be written as as a linear combination

$$|\boldsymbol{\psi}\rangle = \sum_l \psi_l\,|\boldsymbol{\psi}_l\rangle \tag{20}$$

of the eigenstates $|\boldsymbol{\psi}_l\rangle$ of an orthonormal basis. Here, we use the Zeeman basis[18]

$$|\boldsymbol{\psi}_l\rangle = \left|j_1^l j_2^l \dots j_N^l\right\rangle \tag{21}$$

with the nth spin ($n \in \{1,2,\dots,N\}$) in the lth configuration: $j_n^l \in \{-S,-S+1,\dots,S-1,S\}$; S is the spin quantum number of the nth spin. Then, the time dependence is given by the time-dependent Schrödinger equation. Due to the fact that the eigenstates $|\boldsymbol{\psi}_l\rangle$ are time independent, we get a matrix for the expansion coefficients ψ_l:

$$i\hbar\frac{\partial\psi_n}{\partial t} = \hat{\boldsymbol{\mathcal{H}}}\psi_n \tag{22}$$

with the matrix elements

$$\hat{\mathcal{H}}_{lm} = \langle\boldsymbol{\psi}_l|\hat{\mathcal{H}}|\boldsymbol{\psi}_m\rangle \tag{23}$$

of the Hamilton operator matrix $\hat{\boldsymbol{\mathcal{H}}}$. The expansion coefficients ψ_l themselves are complex numbers:

$$\psi_n = \psi_n^{\mathrm{Re}} + i\psi_n^{\mathrm{Im}}. \tag{24}$$

Therefore, we can split the matrix equation (22) in two coupled matrix equations for ψ_n^{Re} and ψ_n^{Im}:

$$\frac{\partial\psi_n^{\mathrm{Re}}}{\partial t} = \frac{1}{\hbar}\hat{\boldsymbol{\mathcal{H}}}\psi_n^{\mathrm{Im}}, \tag{25a}$$

$$\frac{\partial\psi_n^{\mathrm{Im}}}{\partial t} = -\frac{1}{\hbar}\hat{\boldsymbol{\mathcal{H}}}\psi_n^{\mathrm{Re}}, \tag{25b}$$

which have been solved numerically. We are interested in the time dependence of all spins separately. Therefore, we have calculated the expectation values

$$\langle\hat{S}_n^{\alpha}\rangle = \langle\boldsymbol{\psi}|\hat{S}_n^{\alpha}|\boldsymbol{\psi}\rangle \tag{26}$$

for each spin n.

For comparison, we have also solved the Landau-Lifshitz equation

$$\frac{\partial\mathbf{S}_n}{\partial t} = \frac{\gamma}{\mu_S}\mathbf{S}_n \times \mathbf{H}_n \tag{27}$$

with the effective fields $\mathbf{H}_n = -\mathrm{grad}_{\mathbf{S}_n}\mathcal{H}$ and calculated the time dependence of S_n^{α} ($\alpha \in \{x,y,z\}$). The prefactor γ/μ_S comes from the relation of the magnetic moment of the electron to the related spin moment.[19] Due to this prefactor, different time scales in quantum mechanical and classical descriptions occur. Here, we are interested in the relation between the expectation values of the quantum mechanical spin and the classical spin. Therefore, we set $\hbar = \gamma = \mu_S = 1$.

A. Single atom

In the following, we describe a simple single-spin model. Such a model can be seen as a prototype for a single atom on a nonmagnetic surface[20,21] or as a model for a simple magnetic molecule. The corresponding Hamiltonian H (Hamilton operator $\hat{\mathcal{H}}$ / Hamilton function $\mathcal{H}$) is given by

$$\mathsf{H} = -D_z(\mathsf{S}_z)^2 - \mu_S B_z \mathsf{S}_z - \mu_S B_x(t)\mathsf{S}_x. \tag{28}$$

Here, we use a slightly different notation as before. Hence, $\mathsf{S}_n = (\mathsf{S}_n^x,\mathsf{S}_n^y,\mathsf{S}_n^z)$ is either the classical spin $\mathbf{S}_n$ or the spin operator $\hat{\mathbf{S}}_n$.

The first term of the Hamiltonian describes a uniaxial anisotropy with the z axis as the easy axis. The second term represents a static external magnetic field in the $+z$ direction. The last term represents a time-dependent field pulse $B_x(t) = B_0^x e^{-\frac{1}{2}\left(\frac{t-t_0}{T_W}\right)^2}$ with Gaussian shape to excite the spin. In an experimental setup using single atoms, such an excitation can be realized, e.g., by a current pulse coming from a scanning tunneling microscope (STM) tip. In the following, we investigate the two situations that either (i) $D_z = 0$ and $B_z \neq 0$ or, vice versa, (ii) $D_z \neq 0$ and $B_z = 0$. In the case (i), all terms of the Hamiltonian are linear in S, which corresponds to a Heisenberg equation in the form of Eq. (3). In the second case (ii), the Hamiltonian contains a quadratic term and hence the corresponding Heisenberg equation contains a correction term of the order of $\hbar$ [see discussion around Eq. (4)].

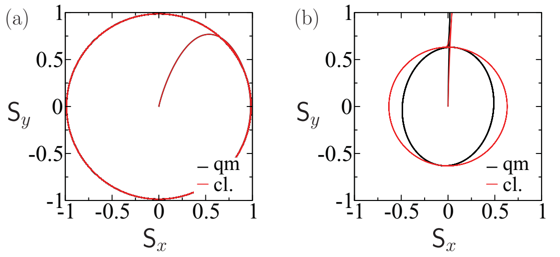

FIG. 1. (Color online) Projection of the spin-space trajectories and S_z versus time t of a single spin described by quantum mechanics (qm) and classical physics (cl.). The assumed parameters are (a) $D_z = 0$, $\mu_S B_z = 2.1$ and (b) $D_z = 0.1$, $\mu_S B_z = 0$. In the quantum mechanical description, we have assumed $S = 1$.

Figure 1 shows the projection of the spin-space trajectory into the S_x-S_y plane. In the initial configuration, the spin was oriented in the $+z$ direction. At $t = t_0 = 10$, the pulse field is switched on with a field strength of $\mu_S B_0^x = 3.27$ and a pulse width $T_W = 0.3$. After the field pulse is gone, the spin precesses on a stable orbit around the effective field. Figure 1(a) shows the situation with the external field oriented in the $+z$ direction ($\mu_S B_z = 2.1$). In this case, we find a perfect match of classical and quantum mechanical trajectories. In both descriptions (classical and quantum), the expected circular orbit and the precession frequency given by the Larmor frequency $\omega = \mu_S B_z$ can be seen. The situation changes drastically if $D_z \neq 0$ (in the following, $D_z = 0.1$). As expected with respect to the discussion in the Sec. I, we do not see an agreement between classical and quantum mechanical trajectories. The classical calculation shows the expected circular orbit. However, the quantum mechanical calculation shows an elliptical orbit for the expectation value $\langle\hat{\mathbf{S}}\rangle$. The ellipticity increases with decreasing $\langle\hat{S}_z\rangle$. For $\langle\hat{S}_z\rangle = 0$, one finds a periodic oscillation of $\langle\hat{S}_y\rangle$ between -1 and $+1$, while $\langle\hat{S}_x\rangle$ is zero for all times. The further decrease of $\langle\hat{S}_z\rangle$ from 0 to -1 leads to a decrease of the ellipticity down to zero again.

Concerning the precession frequency, the frequency is given in the classical as well as in the quantum mechanical case by Eq. (18) just by setting $J = 0$. Therefore, in the classical description, the frequency is given by $\omega = 2D_z S_z$, which means that there is a strong dependence on S_z: $S_z = 0$ means no precession. Physically, this can be explained due to the degeneracy of the orientation of the effective field. In the quantum mechanical description, the frequency is given by $\omega = 2D_z(S - \frac{1}{2})$. In this description, the frequency only depends on the anisotropy constant D_z and on the spin quantum number S and not on $\langle\hat{S}_n^z\rangle$. This also means that, in the case $\langle\hat{S}_n^z\rangle = 0$, the frequency is still $\omega = 2D_z(S - \frac{1}{2})$. Figure 2 shows S_x as function of time. In all cases, the curves can be fitted by a sine or cosine with the expected frequencies discussed before.

B. Trimer

In the preceding section, we have described the investigations of a single spin. To investigate the influence of an exchange interaction, we now look for three coupled spins in a row. In reality, such a configuration corresponds with a trimer (three atoms in a row).[22,23] The Hamiltonian H (Hamilton operator $\hat{\mathcal{H}}$ / Hamilton function $\mathcal{H}$),

$$\mathsf{H} = -J\sum_{\langle l,m\rangle} \mathsf{S}_l\cdot\mathsf{S}_m - D_z\sum_l \left(\mathsf{S}_l^z\right)^2 - \mu_S B_z\sum_l \mathsf{S}_l^z - \mu_S B_0^x e^{-\frac{1}{2}\left(\frac{t-t_0}{T_W}\right)^2}\mathsf{S}_1^x, \qquad (29)$$

has slightly changed by adding the sum term describing the ferromagnetic exchange interaction between nearest neighbors (spin 1 with spin 2 and spin 2 with spin 3; we assumed no coupling between the spins 1 and 3). Further, the field pulse is only acting on the first atom. Apart from that, we have used the same notation as in the preceding section.

For the trimer, we have performed similar calculations as before: $B_z = 0.1, D_z = 0$ and $B_z = 0, D_Z = 0.1$. In both cases, we have assumed $J = 1$. The upper row of Fig. 3 shows the projection of the trajectories $\langle\hat{\mathbf{S}}_n\rangle$ of all three spins $n \in \{1,2,3\}$ for the latter case $B_z = 0$ and $D_z = 0.1$. The lower row shows the corresponding trajectories in the classical spin space. As expected, there is a deviation between the classical and the quantum mechanical trajectories. Furthermore, the difference between frequencies in the quantum mechanical $\omega = 2D_z(S - \frac{1}{2})$ and the classical $\omega = 2D_z S_z$ descriptions can be clearly seen by the bisection of the number of lines in the lower figures. To clarify the role of the exchange interaction, we also calculated the trajectories for $B_z = 0.1$ and $D_z = 0$. Figure 4 shows S_n^x

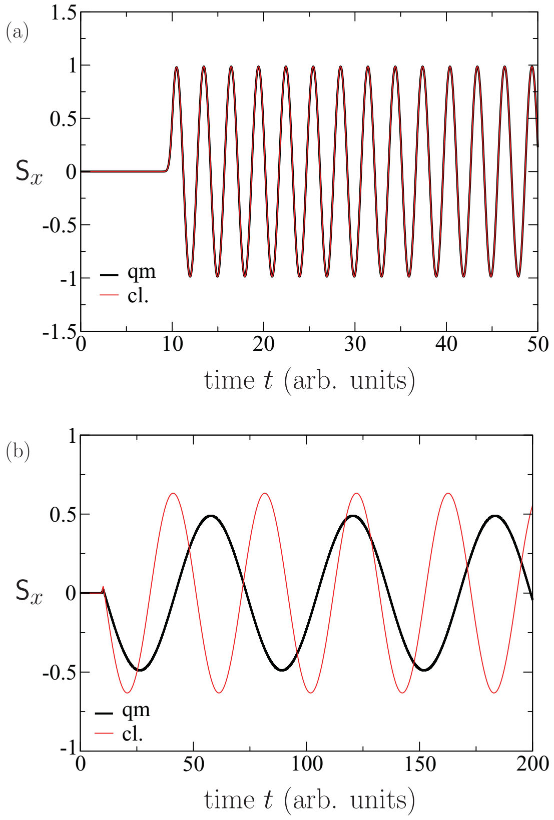

FIG. 2. (Color online) Time evolution of the quantum mechanical (qm) expectation value $\mathsf{S}_x = \langle\hat{S}_x\rangle$ for $S = 1$ and the x component of a classical (cl.) spin $\mathsf{S}_x = S_x$. The curves correspond with the trajectories of Fig. 1: (a) $D_z = 0$, $\mu_S B_z = 2.1$ and (b) $D_z = 0.1$, $\mu_S B_z = 0$.

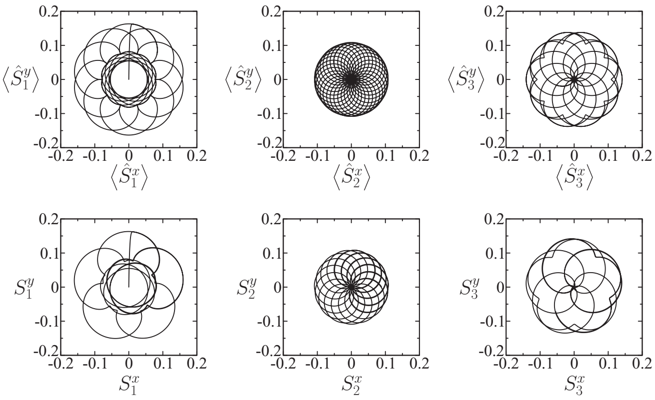

FIG. 3. Projected trajectories of the three atoms of a trimer (from left to right). Upper row: trajectories from the quantum mechanical expectation value $\langle \hat{\mathbf{S}}_n \rangle$; lower row: the trajectories from the corresponding classical spins $\mathbf{S}_n$ ($n \in \{1,2,3\}$). The assumed parameters are $D_z = 0.1$, $\mu_S B_z = 0$, and $S = 1$ in the quantum mechanical description.

and $\langle \hat{S}_n^x \rangle$ as a function of time. We see a perfect match between the classical S_n^x and the quantum mechanical expectation value $\langle \hat{S}_n^x \rangle$. We could expect this behavior due to the linearity of the exchange interaction concerning $\hat{\mathbf{S}}_n$ (see discussion in Sec. I).

IV. ADDITIONAL CONSIDERATIONS

The previous results can be brought in broader context. In literature, one can find similar results or hints. However, in most of these cases, the results are by-products and no detailed discussion can be found. Furthermore, in some cases, the publications are describing wrong physics. A prominent case can be found in the description of the second quantization (Holstein-Primakoff or Dyson-Maleev). Most of the papers dealing with single-ion anisotropy are using a classical instead of the correct quantum mechanical description. Carelessness during the calculation directly leads to the classical result $2D_z S\hbar^2 a^+ a$.

The starting point is the Hamiltonian of the single-ion anisotropy

$$\hat{\mathcal{H}}_{D_z} = -D_z \sum_l \hat{S}_l^z \hat{S}_l^z. \tag{30}$$

With the transformation

$$\hat{S}_z = \hbar(S - a^+ a), \tag{31}$$

we get directly

$$\hat{\mathcal{H}}_{D_z} = -D_z \hbar^2 \sum_l [S^2 - 2S a_l^+ a_l + a_l^+ a_l a_l^+ a_l]. \tag{32}$$

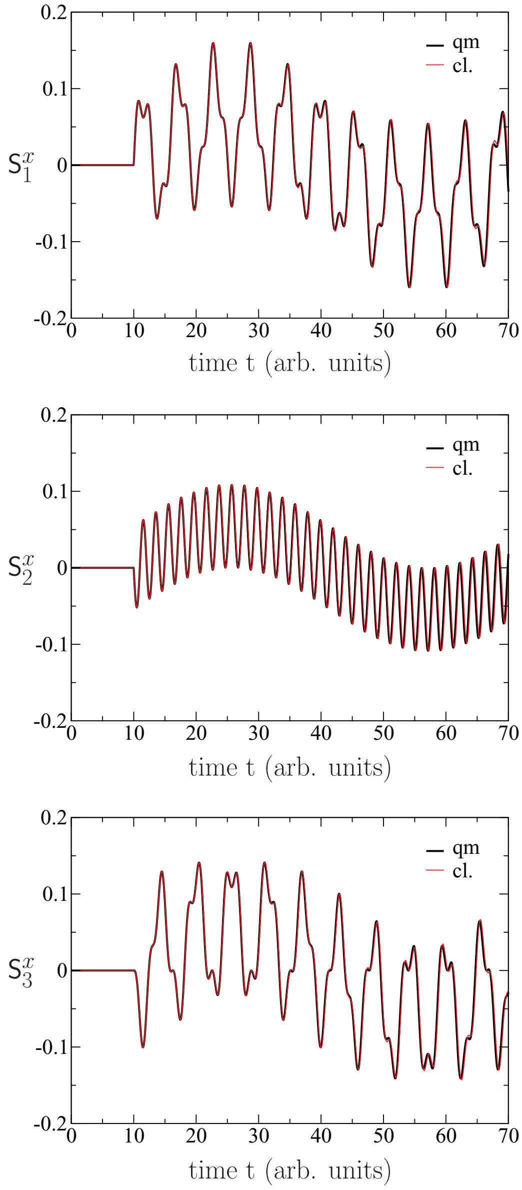

FIG. 4. (Color online) Time evolution of the quantum mechanical (qm) expectation values $\mathsf{S}_n^x = \langle \hat{S}_n^x \rangle$ ($n \in \{1,2,3\}$) and the classical x components $\mathsf{S}_n^x = S_n^x$ of a trimer. The assumed parameters are $D_z = 0$, $\mu_S B_z = 0.1$, and $S = 1$ in the quantum mechanical description.

The first term of the sum on the right-hand side gives the constant ground-state energy. The second-order term $a_l^+ a_l$ describes the excitation of linear spin waves, while the fourth-order term describes the interaction between two spin waves. In the following, we are not interested in such interactions, which means that we skip in the following all fourth-order terms. However, if we skip $a_l^+ a_l a_l^+ a_l$ at this point, we end up with the wrong classical solution

$$\hat{\mathcal{H}}_{D_z}^{\text{cl.}} = 2D_z S\hbar^2 \sum_l a_l^+ a_l \tag{33}$$

because $a_l^+ a_l a_l^+ a_l$ is a mixture of the second-order term $a_l^+ a_l$ and the truly fourth-order term $a_l^+ a_l^+ a_l a_l$:

$$\begin{aligned} [a_l, a_l^+] &= a_l a_l^+ - a_l^+ a_l = 1 \\ \Rightarrow a_l^+ a_l a_l^+ a_l &= a_l^+ a_l + a_l^+ a_l^+ a_l a_l. \end{aligned} \tag{34}$$

After skipping the correct fourth-order term, we end up with

$$\hat{\mathcal{H}}_{D_z} = \hbar^2 \sum_l \left[-D_z S^2 + 2D_z \left(S - \frac{1}{2} \right) a_l^+ a_l \right]. \quad (35)$$

The extension of the Vaks-Larkin-Pikin (VLP) diagrammatic technique to the case of single-ion anisotropies is a further example where carelessness leads to serious difficulties.[24]

Another method where one can find the correct energy contribution of the single-ion anisotropy $E_{D_z} = 2D_z(S - \frac{1}{2})$ is the Green's function formalism of the quantum Heisenberg model. In contrast to the exchange interaction, the contribution of single-ion anisotropy to the Green's function can not be decoupled by the RPA (random phase approximation) decoupling. In these cases, the Callen-Anderson decoupling leads to the correct results[25]:

$$D_z \langle\langle \hat{S}_l^+ \hat{S}_l^z + \hat{S}_l^z \hat{S}_l^+ ; \left(\hat{S}_m^z\right)^n \hat{S}_m^- \rangle\rangle \to D_z \Phi^+ G_{lm}^+ \quad (36)$$

with the decoupled Green's function

$$G_{lm}^+ = \langle\langle \hat{S}_l^+ ; \left(\hat{S}_m^z\right)^n \hat{S}_m^- \rangle\rangle \quad (37)$$

and

$$\Phi^+ = 2\langle \hat{S}_l^z \rangle \left(1 - \frac{1}{2S^2} \left[S(S+1) - \langle (\hat{S}_l^z)^2 \rangle \right] \right). \quad (38)$$

This decoupling holds for all temperatures T. However, for comparison, we are interested in the limit $T \to 0$. In this limit, the expectation value $\langle \hat{S}_l^z \rangle$ becomes S and the correlation $\langle (\hat{S}_l^z)^2 \rangle \to S^2$. Therewith, we get again

$$E_{D_z} = D_z \Phi^+ = 2D_z \left(S - \frac{1}{2} \right). \quad (39)$$

This result was first derived in the context of double-time Green's functions by Kazakov in 1977.[26]

The last example we discuss is the description using coherent states as a wave-function basis set. In this description, the corresponding Heisenberg equation looks like the classical Landau-Lifshitz equation in spherical coordinates (see Balakrishnan *et al.*[27]) with just one difference: the prefactor of the anisotropy term is $D_z\,(2S - 1)$ instead of $2D_z S$ as in the Landau-Lifshitz equation. For a thorough discussion between the classical and the quantum descriptions in the context of coherent states, see also Balakrishnan and Bishop.[28]

In all the cases mentioned before (except the VLP diagrammatic technique), the same contribution of the single-ion anisotropy $2D_z(S - \frac{1}{2})$ can be found. Unfortunately, in the literature, different descriptions can be found, mostly the wrong classical $2D_z S$ one.

V. SUMMARY

In summary, we have shown that the textbook description of the derivation of the Landau-Lifshitz equation starting with the quantum mechanical Heisenberg equation is correct but incomplete. As textbooks confirm, terms linear in $\hat{\mathbf{S}}_n$ in the Heisenberg model indeed lead to an equation similar to the Landau-Lifshitz equation. However, if there are terms quadratic in $\hat{\mathbf{S}}_n$ as the single-ion anisotropy, the situation changes. In this case, an additional term of the order of $\hbar$ occurs. Alternatively, the occurring equation can be written as two but mirrored Landau-Lifshitz equations [see Eq. (5)] due to the noncommutativity of $\hat{\mathbf{S}}_n$ with the effective field $\hat{\mathbf{H}}_n$ in this case. In the end, this means that the dynamics of a system that can be described with a Heisenberg model containing only linear terms in $\hat{\mathbf{S}}_n$ (such as soft-magnetic materials, e.g., permalloy) obey classical laws. Therefore, the classical description leads to correct physical results. However, quantum mechanical systems described by a Heisenberg model with quadratic terms such as the single-ion anisotropy can not be described classically. Here, we need the full quantum mechanical description.

In a wider context, this behavior is nothing other than the known failure of the Ehrenfest theorem in the case of nonlinear potentials $\mathrm{U}(\hat{\mathbf{r}})$. Here, we have the analogies between the force $\hat{\mathbf{F}}$ and the effective field $\hat{\mathbf{H}}$ as well as between the potential $\mathrm{U}(\hat{\mathbf{r}})$ and Hamiltonian $\hat{\mathcal{H}}(\hat{\mathbf{S}})$:

$$\hat{\mathbf{F}} = -\mathrm{grad}_{\hat{\mathbf{r}}} \mathrm{U}(\hat{\mathbf{r}}) \leftrightarrow \hat{\mathbf{H}} = -\mathrm{grad}_{\hat{\mathbf{S}}} \hat{\mathcal{H}}(\hat{\mathbf{S}}).$$

The description given within this publication is not restricted to ferromagnets. Antiferromagnets as well as ferrimagnets show the same behavior as long as there is no relaxation process. It is known that the quantum mechanical ground state of an antiferromagnet is unequal to the classical Néel state. Therefore, the relaxation to the ground state shows different trajectories depending on the description being classical or quantum mechanical. The same is true for ferrimagnets. In the case of a ferromagnet, the relaxation does not change the situation caused by the fact that the classical and quantum mechanical ground states are equal.

In the second part of this paper, we have proved our statement analytically and with numerical methods. The systems we investigated were described by Heisenberg Hamiltonians with exchange, external field, and single-ion anisotropies. If we skip the anisotropy term, a perfect match between classical description and quantum mechanics occur. However, if the anisotropy is taken into account, we only find similarities between both descriptions, but no agreement. The essence of these calculations is the change from the S_z-dependent classical frequency $\omega = 2D_z S_z$ to the quantum mechanical frequency $\omega = 2D_z(S - \frac{1}{2})$, which only depends on the spin quantum number S and the anisotropy constant D_z. This frequency can be found for several physical descriptions and models, unfortunately with less paid attention in literature.

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APPENDIX: FERROMAGNETIC SPIN WAVES (SOLVING THE SCHRÖDINGER EQUATION)

The Schrödinger equation

$$i\hbar \frac{\partial \left| \psi \right\rangle}{\partial t} = \hat{\mathcal{H}} \left| \psi \right\rangle \quad (A1)$$

with Hamiltonian

$$\hat{\mathcal{H}} = -\frac{1}{2\hbar^2}\sum_{l,m} J_{lm}\left[\hat{S}_l^z\hat{S}_m^z + \frac{1}{2}(\hat{S}_l^+\hat{S}_m^- + \hat{S}_l^-\hat{S}_m^+)\right] - \frac{D_z}{\hbar^2}\sum_l \left(\hat{S}_l^z\right)^2 \tag{A2}$$

can be solved by the following wave ansatz, which describes the excitation of one magnon:

$$|\psi\rangle = \frac{1}{\hbar\sqrt{2NS}}\sum_n e^{i(\mathbf{k}\cdot\mathbf{r}_n-\omega t)}\hat{S}_n^-\,|\psi_0\rangle\,, \tag{A3}$$

where $|\psi_0\rangle$ is the ground-state configuration where all spins are oriented in the $+z$ direction.

With this ansatz, the left-hand side of the Schrödinger equation (A1) becomes

$$i\hbar\frac{\partial\,|\psi\rangle}{\partial t} = \hbar\omega\,|\psi\rangle\,. \tag{A4}$$

In order to get the energy $\langle\hat{\mathcal{H}}\rangle = \langle\psi|\,\hat{\mathcal{H}}\,|\psi\rangle = \hbar\omega$ on the right-hand side of Eq. (A1), we have to evaluate

$$\begin{aligned}\hat{\mathcal{H}}(\hat{S}_n^-\,|\psi_0\rangle) &= \hat{S}_n^-\hat{\mathcal{H}}\,|\psi_0\rangle + [\mathcal{H},\hat{S}_n^-]\,|\psi_0\rangle\\ &= E_0\hat{S}_n^-\,|\psi_0\rangle + [\hat{\mathcal{H}},\hat{S}_n^-]\,|\psi_0\rangle\,.\end{aligned} \tag{A5}$$

E_0 is the ground state energy: $\hat{\mathcal{H}}\,|\psi_0\rangle = E_0\,|\psi_0\rangle$. Further, the commutator $[\hat{\mathcal{H}},\hat{S}_n^-]$ is given by

$$[\hat{\mathcal{H}},\hat{S}_n^-] = \sum_l \frac{J_{ln}}{\hbar}\left(\hat{S}_l^-\hat{S}_n^z - \hat{S}_n^-\hat{S}_l^z\right) + 2\frac{D_z}{\hbar}\left(\hat{S}_l^z - \frac{\hbar}{2}\right)\hat{S}_n^-.$$

Therewith, the right-hand side of Eq. (A1) becomes, with $\hat{S}_l^z\,|\psi_0\rangle = S\hbar\,|\psi_0\rangle$,

$$\begin{aligned}\hat{\mathcal{H}}\,|\psi\rangle &= E_0\,|\psi\rangle\\ &\quad+\frac{1}{\hbar\sqrt{2NS}}\left\{\sum_{l,n} J_{ln}Se^{i(\mathbf{k}\cdot\mathbf{r}_n-\omega t)}(\hat{S}_n^- - \hat{S}_l^-)\right.\\ &\quad\left.+\,2D_z\left(S-\frac{1}{2}\right)\sum_n e^{i(\mathbf{k}\cdot\mathbf{r}_n-\omega t)}\hat{S}_n^-\right\}|\psi_0\rangle\,.\end{aligned}$$

Next, one has to exchange the two summation indices in such a way that $\hat{S}_l^-$ is labeled as $\hat{S}_n^-$ and exploit the symmetry $J_{nl} = J_{ln}$:

$$\begin{aligned}&\sum_{l,n} J_{ln}Se^{i(\mathbf{k}\cdot\mathbf{r}_n-\omega t)}(\hat{S}_n^- - \hat{S}_l^-)\\ &\quad= \sum_n\sum_l J_{ln}Se^{i(\mathbf{k}\cdot\mathbf{r}_l-\omega t)}\hat{S}_l^-\\ &\qquad-\sum_l\sum_n J_{nl}Se^{i(\mathbf{k}\cdot\mathbf{r}_n-\omega t)}\hat{S}_l^-\\ &\quad= \sum_{l,n} J_{ln}(1-e^{i\mathbf{k}\cdot(\mathbf{r}_l-\mathbf{r}_n)})e^{i(\mathbf{k}\cdot\mathbf{r}_n-\omega t)}\hat{S}_n^-.\end{aligned}$$

With this transformation, we get

$$\begin{aligned}\hat{\mathcal{H}}\,|\psi\rangle &= E_0\,|\psi\rangle + \frac{1}{\hbar\sqrt{2NS}}\sum_n\left\{\sum_l J_{ln}S(1-e^{i\mathbf{k}\cdot(\mathbf{r}_l-\mathbf{r}_n)})\right.\\ &\quad\left.+\,2D_z\left(S-\frac{1}{2}\right)\right\}e^{i(\mathbf{k}\cdot\mathbf{r}_n-\omega t)}\hat{S}_n^-\,|\psi_0\rangle\,.\end{aligned} \tag{A6}$$

By putting Eqs. (A6) and (A4) (left- and right-hand sides of the Schrödinger equation) together, we get the spin-wave energy by setting $E_0 = 0$:

$$\hbar\omega = S[J(0)-J(k)] + 2D_z\left(S-\frac{1}{2}\right). \tag{A7}$$

Here, we have used the abbreviation (Fourier transformation of the exchange constant)

$$J(k) = \sum_l J_{ln}e^{i\mathbf{k}\cdot(\mathbf{r}_l-\mathbf{r}_n)}. \tag{A8}$$

Assuming only nearest-neighbor exchange interaction, $J(0) - J(k)$ becomes $2J[1-\cos{(ka)}]$ and the spin-wave energy

$$\hbar\omega = 2SJ[1-\cos{(ka)}] + 2D_z\left(S-\frac{1}{2}\right). \tag{A9}$$

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