PHYSICAL REVIEW B VOLUME 60, NUMBER 21 1 DECEMBER 1999-I

Dynamic equations for individual spins in materials with magnetic order: The ideal ferromagnet

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(Received 24 March 1999)

The time and temperature dependence of individual spins is studied in an ideal ferromagnet. Starting from the Heisenberg Hamiltonian in a magnetic field and, building on linear-response theory, we derive the linearized microscopic analogy to the macroscopic Landau-Lifshitz equations of motion for magnetization. The dynamical equations take into account the influence of spin correlations in addition to the molecular field. In order to illustrate the validity of our approach, we calculate the temperature-dependent spin-wave spectrum and Landau damping by employing a finite-temperature perturbation theory for spin operators. The latter is adapted to suit higher-order spin-correlation functions which can be described by simple quasiparticle interactions with the spin system. Our results agree with those established on the thermodynamics of the Heisenberg model. In addition, we provide a method of how to deduce macroscopic equations of the Landau-Lifshitz type from first principles, including temperature dependence and damping. [S0163-1829(99)06141-X]

I. INTRODUCTION

An effective way to find out the characteristic properties of ferromagnetic materials is to study their specific response towards a magnetic field. A relation that describes the connection between this field and the magnetization of the material is provided by the macroscopic Landau-Lifshitz equations of motion.[1] However, in past years experimental work advanced to the limits of macroscopic physics when investigating magnetic structures. This led, for example, to the observation of exchange-dominated spin-wave resonance spectra and traveling spin waves in ferrite films[2–4] or the magnetization reversal of single-domain particles which might serve as magnetic memory elements.[5–8] The former effect is based on exciting very short spin waves, whereas the latter on the smallness of the structure. On the other hand, the theoretical approach to the problem of magnetization dynamics still remains founded essentially on the theory of Landau and Lifshitz, i.e., on a macroscopic model. Although this model gives a good understanding of the underlying physics, a natural limit is set by this model to a sufficient description of the experiments when using ultrasmall wavelengths or sizes.

This paper deals with the development of dynamic equations for the average of individual spins in magnetic ordered materials or, in a sense, with the derivation of a microscopic analogy to the macroscopic Landau-Lifshitz equations. However, there are significant differences to the classical treatment. As our approach is a microscopic one, it is possible to obtain all material properties, described by various magnetic susceptibilities and the equilibrium magnetization, from basic principles. What is more important is that individual spins become correlated. This happens as a time-dependent external magnetic field drives the system out of equilibrium, by exciting the individual spins. In this way, the spins interact with each other, not only as if each of them is surrounded by an average Weiss molecular field, as in the classical case, but in a way that takes into account the nonequilibrium spin correlations. These correlations give rise to effects such as the damping of the motion of the spin system and the temperature dependence of the dynamic parameters. They also introduce memory effects on how the system was perturbed by the external field.

A brief formulation of the problem was communicated before together with some preliminary results.[9] Here, we develop this approach in a more systematic manner. To keep the calculations simple and focus on the main aspects of the theory, we restrict our analysis to an ideal Heisenberg ferromagnet. In the first part of the paper the basic equations of motion for the average of individual spins are introduced. Their structure is a hierarchy, allowing, for example, to separate single spin-wave excitations from spin-wave correlations. In the second part, we address the following two questions: Does our approach allow one to obtain previously established results on the thermodynamics of the Heisenberg model; and is it possible to derive from the microscopic equations macroscopic equations for the magnetization, still incorporating such effects as temperature dependence and damping? By applying a finite-temperature diagrammatic perturbation theory, solutions to these problems are provided for the example of an ideal ferromagnet. The perturbation theory builds on the diagram-technique developed by Izyumov, Kassan-Ogly, and Skryabin.[10,11] Here, we adapt it to deal with more complicated three-spin correlation functions as they arise in the derivation of the equations of motion. In principle, this theory may be used to describe the dynamics of individual spins for all temperatures.[11] However, in order to make contact with earlier treatments on the thermodynamics of the Heisenberg model, in particular with those of Stinchcombe *et al.*[12] as well as Vaks, Larkin, and Pikin,[13,14] we exclude the region very close to the critical temperature T_c of magnetic ordering and examine the large range of temperatures, where the principle features of ferromagnetism are pronounced rather than those of phase transition.

In subsequent publications we will develop our approach further to describe more realistic systems such as magnetic multilayers and single-domain particles. In particular, the dynamic equations will include apart from exchange also dipole-dipole interaction and anisotropy effects. Extensions to include antiferromagnetic and ferrimagnetic crystals are also planned. Based on these dynamic equations, we hope that it will be possible to solve many outstanding problems in the field of magneto and nanoelectronics.

Previously, different attempts were made to develop the theory of dynamic properties in magnetic ordered systems, for example, using the Lagrangian[15] or Hamiltonian[16] formalism, by obtaining dynamic equations from simple symmetry considerations,[17] or in the form of hydrodynamic equations of spin waves.[18] However, the obtained nonlinear equations still retain the form of the original formulation by Landau and Lifshitz. Besides their macroscopic nature, the main disadvantage of these approaches is that damping and temperature dependence cannot be derived from basic principles, so that they are at best just added as phenomenological terms.

On the other hand, it is possible to solve the dynamics of an individual spin based purely on quantum-mechanical considerations such as the diagonalization of the respective Hamiltonian[19] or solving the equation of motion for spin operators.[20] Unfortunately, these approaches neglect the thermal fluctuations of the system which are often of importance.

A way around this problem are thermodynamic considerations on the magnetic materials, in particular, using quantum statistical mechanics. Although quantum statistical mechanics allow for a broad range of powerful methods to yield a proper description of magnetic materials, most of the obtained results have been restricted to rather ideal situations.[12–14,21] Some interesting generalizations of these approaches to the solution of more realistic problems have been made, for example, by Erickson and Mills within the context of ultrathin ferromagnetic films as they occur in multilayer magnetic materials.[22] Nevertheless, when dealing with particular boundary problems, the Landau-Lifshitz equations remain the only universal and effective approach to the description of magnetization dynamics. Our aim, therefore, is to compromise between the different views and to advance to dynamic equations for individual spins that combine some of the universal features of the macroscopic approach and the rigor of the quantum statistical formulation. In the macroscopic limit, we obtain equations of the Landau-Lifshitz-type, however, with the important addition that spin correlations are still present, thus meeting the proposed objectives.

The remainder of the paper is structured as follows: In Sec. II we introduce and discuss the simple model of an ideal ferromagnet. Section III is devoted to a general derivation of the dynamic equations for individual spin averages which are then linearized and discussed in some detail. The equations of motion are shown to represent a closed set of differential equations, so that knowing the equilibrium properties of the system, any dynamical property of the system can be calculated even in the close vicinity of T_c. The diagrammatic perturbation theory for spin operators is laid out in Sec. IV. There, we also calculate the three-spin correlation functions which are described by simple quasiparticle interactions of the spin system. These functions are used to describe the correlations in the dynamics of an ideal Heisenberg ferromagnet in Sec. V. Further, we discuss briefly the temperature dependence of the corresponding spectrum which agrees with previous solutions in the limit of an uniform excitation in an infinite lattice. What is more important, we show how to obtain macroscopic equations of motion for the magnetization that depend on temperature and include damping. In conclusion, we summarize our results and point out further directions in Sec. VI.

II. MODEL OF AN IDEAL FERROMAGNET

In the present paper we consider the case of an ideal ferromagnet where the Hamiltonian $H=H_{\text{ex}}+H_{\text{z}}$ is a sum of contributions arising from the quantum-mechanical exchange interaction and the Zeeman energy. For our purposes, we choose the assumption of localized magnetic moments at the lattice sites, where the exchange interaction between them is described by the Heisenberg Hamiltonian,

$$H_{\text{ex}}=-\tfrac{1}{2}\sum_{i,j}{}' \, I_{ij}\mathbf{S}_i\cdot\mathbf{S}_j=-\tfrac{1}{2}\sum_{i,j}{}' \, I_{ij}(S_i^-S_j^+ + S_i^zS_j^z). \tag{1}$$

The prime indicates that the sum is only taken at different lattice sites $i\neq j$ over a product of spin operators S_i^μ and S_j^ν $(\mu,\nu=+,-,z)$ which obey the following commutator relations:

$$[S_i^+,S_j^-]=2\delta_{ij}S_i^z;\quad [S_i^z,S_j^\pm]=\pm\delta_{ij}S_i^\pm. \tag{2}$$

The coefficients $I_{ij}=I_{ji}$ are the exchange integrals. Besides neglecting contributions such as dipole-dipole interaction or anisotropy effects, the assumption of localized moments is a gross simplification of the physical reality. In order to describe magnetic materials and structures more accurately, their full electron system has to be taken into account. Nevertheless the Heisenberg model is thought to be suitable for many ferromagnetic dielectrics and semiconductors, whereas it is less satisfactory for metallic ferromagnets where the itinerant contribution of the conduction electrons is significant. If, however, as is standard practice, the exchange integrals are not calculated from basic principles but taken from experiments, the experimental value will already contain the itinerant electron contribution and the localized electron picture is still a reasonable approximation for a wide range of metals with magnetic order.[23] This is in particular the case for the rare earths which have a specialized form of I_{ij} due to the indirect coupling of the localized f electrons through the conduction band. The second term in the Hamiltonian relates to the Zeeman energy,

$$H_{\text{z}}=g\mu_B\sum_i \mathbf{h}_i(t)\cdot\mathbf{S}_i=g\mu_B\sum_i \{\tfrac{1}{2}(h_i^-(t)S_i^+ + h_i^+(t)S_i^-) + h_i^z(t)S_i^z\}, \tag{3}$$

and describes the interaction of the localized moments with an external time-dependent magnetic field $\mathbf{h}_i(t)=\mathbf{h}(\mathbf{r}_i,t)$ with cyclic components defined in analogy to the usual definition for spin operators, i.e., $h_i^\pm(t)=h_i^x(t)\pm ih_i^y(t)$. The Bohr magneton is defined as $\mu_B=|e|\hbar/(2m_ec)$, and g is the Landé factor.

III. DYNAMIC EQUATIONS FOR AN INDIVIDUAL SPIN AVERAGE

In order to study the time and temperature dependence of individual spins embedded in a ferromagnetic crystal, we average the spin operators over the nonequilibrium statistical ensemble of the entire system,

$$\langle \mathbf{S}_i \rangle = \mathrm{Sp}\{\rho \mathbf{S}_i\}, \tag{4}$$

where Sp denotes the trace. The density operator $\rho(t)$ contains the complete information about the ensemble. It is thus sufficient to use the von Neumann equation of motion for ρ to describe the time development of the system, $i\hbar\, \partial_t \rho = [H,\rho]$. Together with Eq. (4) this leads to an equation for the spin average,

$$i\hbar\, \partial_t \langle \mathbf{S}_i \rangle = \langle [\mathbf{S}_i, H] \rangle. \tag{5}$$

With help of the relations (2) the commutator in Eq. (5) can be calculated. Introducing an operator describing the spin fluctuations $s_i^\mu = S_i^\mu - \langle S_i^\mu \rangle$, we replace Eq. (5) with the following set of equations:

$$i\, \partial_t \langle S_i^+ \rangle = \gamma [h_{i,\mathrm{eff}} \langle S_i \rangle]^{+z} - \hbar^{-1} \mathcal{F}_i^+ , \tag{6a}$$

$$-i\, \partial_t \langle S_i^- \rangle = \gamma [h_{i,\mathrm{eff}} \langle S_i \rangle]^{-z} - \hbar^{-1} \mathcal{F}_i^- , \tag{6b}$$

$$2i\, \partial_t \langle S_i^z \rangle = \gamma [h_{i,\mathrm{eff}} \langle S_i \rangle]^{-+} - \hbar^{-1} \mathcal{F}_i^z . \tag{6c}$$

For convenience, we introduced the following notation in analogy to the definition of a vector product:

$$[h_{i,\mathrm{eff}} \langle S_i \rangle]^{\mu\nu} = h_{i,\mathrm{eff}}^\mu \langle S_i^\nu \rangle - h_{i,\mathrm{eff}}^\nu \langle S_i^\mu \rangle, \tag{7}$$

and an effective magnetic field

$$h_{i,\mathrm{eff}}^\mu(t) = h_i^\mu(t) - \frac{1}{g\mu_B} \sum_{j \neq i} I_{ij} \langle S_j^\mu \rangle, \tag{8}$$

where $\gamma = g\mu_B/\hbar > 0$ is the gyromagnetic ratio. The fluctuations in the spin system correlate the spins with each other. These correlations are contained in the terms

$$\mathcal{F}_i^\sigma = \sum_{j \neq i} I_{ij} (\langle s_i^z s_j^\sigma \rangle - \langle s_i^\sigma s_j^z \rangle), \tag{9a}$$

$$\mathcal{F}_i^z = \sum_{j \neq i} I_{ij} (\langle s_i^+ s_j^- \rangle - \langle s_i^- s_j^+ \rangle), \tag{9b}$$

where $\sigma = +,-$. So far no approximations were made and Eq. (6) are exact. The second terms on the right-hand side of Eqs. (6a)–(6c) figure the contribution of the spin fluctuations. Without the spin correlations (9) the equations of motion (6) would yield a closed system of differential equations, that corresponds formally to the solution for the classical precession of a magnetic moment. It is necessary to investigate these correlation terms further.

A standard procedure would be to continue with the equation of motion technique and write an equation of motion for the averages $\langle s_i^\mu s_j^\nu \rangle$. In general, if we repeat this method, this would lead eventually to an infinite series of equations of motion, which have to be approximated by decoupling them at some stage. Since we are interested in the combined dynamical and statistical properties of the system, we study two types of fluctuations more or less simultaneously: coherent fluctuations created by a time-dependent external magnetic field and noncoherent fluctuations due to spontaneous and thermal excitations of the spin system. Whereas coherent fluctuations exist only in the nonequilibrium state of the system, noncoherent fluctuations are an inherent property of its thermodynamic ground state. This does, however, not imply that they can be regarded as independent of each other. In fact, quite the opposite is the case, and this property will be exploited in the calculations. In contrast to the equation-of-motion technique for Green's functions, the operators in Eq. (6) are not tied to the Heisenberg presentation and thus time independent. Therefore, we can express the nonequilibrium correlations in terms of equilibrium correlations, by linearizing them in the time-dependent magnetic field. To identify the leading-order terms of the equilibrium correlations, we use perturbation theory. The former is dealt with in the following, whereas the latter is discussed in Sec. IV.

A. Linearization of the equations of motion

The experiments under consideration shall measure the linear response to the external time-dependent magnetic field. We can thus restrict ourselves to the calculation of the averages $\langle s_i^\mu s_j^\nu \rangle$ to first order in the perturbing field. By separating the external magnetic field into its static and time-dependent contributions $\mathbf{h}_i(t) = \mathbf{h}_0 + \mathbf{h}_{i,\sim}(t)$, the Hamiltonian of the system is rewritten in terms of its static part $\overline{H}$ and its time-dependent perturbation $V(t)$, i.e., $H = \overline{H} + V(t)$. We assume without loss of generality that the static component of the magnetic field is parallel to the z coordinate such that $\mathbf{h}_0 = (0,0,-|h_0^z|)$. Further, in a linear approximation of Eq. (6) the time-dependent field can couple to the averages $\langle S_i^\mu \rangle$ only in an experimental setup, where it is applied perpendicular to the static external field. Therefore, this more interesting perpendicular case shall be treated here, i.e., $h_{i,\sim}^z = 0$.

Now, we turn to the linearization of the averages $\langle s_i^\mu s_j^\nu \rangle$ and first discuss the equilibrium case, where the oscillating external magnetic field is switched off. In Eq. (9a) the operators $s^\pm$ are unpaired and, therefore, the equilibrium averages vanish. Although in the case of Eq. (9b) these creation and destruction operators are paired, the averages do, nevertheless, not contribute. The reason is that the spin operators are taken at the same time, so that the corresponding physical process are spontaneous temperature-independent quantum fluctuations which are prohibited by the assumption of complete localization of the spin moments, i.e., $i \neq j$. We can thus focus only on the nonequilibrium contributions.

The time dependence of the density matrix can be found by solving the von Neumann equation in linear approximation with respect to the perturbing field $V(t)$. Starting from equilibrium at an infinite early time, when $V(-\infty) = 0$, we switch on $V(t)$ adiabatically. If we keep only terms linear in the perturbing field, the von Neumann equation leads to the solution

$$\rho(t) = \rho_{\mathrm{eq}} - i/\hbar \int_{-\infty}^{t} dt'\, e^{-i\overline{H}(t-t')/\hbar} [V(t'), \rho_{\mathrm{eq}}] e^{i\overline{H}(t-t')/\hbar}, \tag{10}$$

where $\rho_{\mathrm{eq}}=\exp(-\beta\overline{H})/\mathrm{Sp}\{\exp(-\beta\overline{H})\}$ is the Gibbs equilibrium density matrix, and $\beta=1/(k_BT)$ the inverse absolute temperature. When taking the trace for $s_i^{\mu}s_j^{\nu}$ as in Eq. (4), we derive for the nonequilibrium correlations (9),

$$\mathcal{F}_i^{\sigma}(t)=g\mu_B\sum_l \int_{-\infty}^{\infty}dt'\, h^{\sigma}_{l,\sim}(t')\varsigma^{\bar{\sigma}}_{il}(t-t'), \tag{11}$$

where $\sigma\neq\bar{\sigma}$ $(\sigma,\bar{\sigma}=+,-)$. The contribution to the z coordinate vanishes, since we assumed $h^z_{i,\sim}=0$. Further, we introduced the quantity

$$\varsigma^{\bar{\sigma}}_{il}(t-t')=\sum_{j\neq i} I_{ij}[\Gamma^{(r)}_{z\sigma,\bar{\sigma}}(i,j;l;t-t')-\Gamma^{(r)}_{\sigma z,\bar{\sigma}}(i,j;l;t-t')], \tag{12}$$

describing the spin correlations as an equilibrium property of the system in terms of the retarded three-spin Green's functions

$$\Gamma^{(r)}_{\sigma z,\bar{\sigma}}(i,j;l;t-t')=-\frac{i}{2\hbar}\Theta(t-t') \times\mathrm{Sp}\{\rho_{\mathrm{eq}}[s_i^{\sigma}(t)s_j^{z}(t),s_l^{\bar{\sigma}}(t')]\}, \tag{13}$$

and $\Gamma^{(r)}_{z\sigma,\bar{\sigma}}$ which is defined in analogy. The quantity ς^{σ}_{il} takes the role of a susceptibility tensor, because the contributions of the nonequilibrium correlations to Eq. (6) are proportional to the driving of the time-dependent field $h^{\sigma}_{l,\sim}(t')$. This ''correlation'' susceptibility describes the deviation from the classical precession of the average spins due to the influence of the higher spin correlations. In principle, we could almost stop our analysis here, because the only problem that remains to be solved is to calculate ς^{σ}_{il}, for instance, using a modified version of the diagrammatic perturbation theories of Refs. 10–13.

However, we set out to derive the microscopic analogy of the Landau-Lifshitz equations. These microscopic equations ought to be as convenient as the macroscopic Landau-Lifshitz equations when solving different applied problems. In particular, such problems may be concerned with exploring the free oscillations of a spin system, where a time-dependent external field is absent, and one would like to calculate the eigenfrequencies and wave functions incorporating the effect of spin correlations. In such a case, inserting Eq. (11) directly into Eq. (6) would be rather awkward. As a rule, analyzing the eigenoscillations of a system, we are not interested in the question of how they were excited. If one would, however, use Eq. (11) directly, one needs to know the entire history on how the system was driven out of equilibrium. We, therefore, replace $h^{\sigma}_{l,\sim}$ in Eq. (11) in terms of $\langle s_l^{\sigma}\rangle$ first and only then insert it into Eq. (6).

Repeating the steps performed in deriving Eq. (11), we calculate the linear-response result in the perturbing field for the nonequilibrium average $\langle s_l^{\sigma}\rangle$. As a result, we find for the derivative in time,

$$\partial_t\langle s_i^{\sigma}\rangle=g\mu_B\sum_l \int_{-\infty}^{\infty}dt'\, h^{\sigma}_{l,\sim}(t')\,\partial_t K^{(r)}_{\sigma\bar{\sigma}}(i,l;t-t'), \tag{14}$$

where the retarded Green's function $K^{(r)}_{\sigma\bar{\sigma}}(i,l;t-t')=-i/(2\hbar)\,\Theta(t-t')\,\mathrm{Sp}\{\rho_{\mathrm{eq}}[s_i^{\sigma}(t),s_l^{\bar{\sigma}}(t')]\}$ is proportional to the linear magnetic susceptibility tensor of the system. The derivative in time of $K^{(r)}_{+-}$, for example, leads to the relation

$$\sum_j [i\hbar\delta_{ij}\partial_t-\varepsilon_{ij}]K^{(r)}_{+-}(j,l;t-t')=\Phi^{-}_{il}(t-t'). \tag{15}$$

Here, we made use of another spin operator describing fluctuations around the equilibrium average, i.e., $s_i^{\mu}=S_i^{\mu}-\langle S_i^{\mu}\rangle_{\mathrm{eq}}$, to separate the spin-wave-type energies obtained by ε_{ij},

$$\varepsilon_{ij}=\delta_{ij}\left(-g\mu_Bh_0^z+\sum_{l\neq i} I_{il}\langle S_l^z\rangle_{\mathrm{eq}}\right)-I_{ij}\langle S_i^z\rangle_{\mathrm{eq}}, \tag{16}$$

from the spin correlations

$$\Phi^{-}_{il}(t-t')=\langle S_i^z\rangle_{\mathrm{eq}}\,\delta_{il}\delta(t-t')-\varsigma^{-}_{il}(t-t'), \tag{17}$$

expressed by the second term of definition (17) in terms of the three-spin Green's function introduced above. This right-hand side of Eq. (15) can be regarded as a generalized function of intensity including the spin correlations (12). Taking the complex conjugate of Eq. (15), an expression for $K^{(r)}_{-+}$ is obtained as can be seen from its definition. Similarly, the Hermitian conjugate of the right-hand side of Eq. (13) leads to a simple relation between the three-spin Green's functions, e.g. $\Gamma^{(r)}_{z-,+}=(\Gamma^{(r)}_{z+,-})^{\dagger}$. The Hermitian property in the indices + and − of the linear and correlation susceptibility tensor is what one expects for gyrotropic materials such as an ideal ferromagnet. Replacing the Green's function in Eq. (14) with the help of the relation (15), we find the following equation for the perturbing field:

$$h^{+}_{i,\sim}(t)=\frac{1}{g\mu_B\langle S_i^z\rangle_{\mathrm{eq}}}\left\{\sum_j (i\hbar\delta_{ij}\partial_t-\varepsilon_{ij})\langle s_j^{+}(t)\rangle+\mathcal{F}_i^{+}(t)\right\}, \tag{18}$$

which relates the time-dependent field to the spin-wave-type energies and the spin-correlations. In the same way, we derive an analogous equation for the other cyclic component of the time-dependent magnetic field. Now, if we insert Eq. (18) into Eq. (11), and integrate partially over t', an integral equation for the nonequilibrium correlations is derived

$$\mathcal{F}_i^{+}(t)=\sum_{l,l'} \langle S_l^z\rangle_{\mathrm{eq}}^{-1}\int_{-\infty}^{\infty}dt'\,\{\langle s_l^{+}(t')\rangle(-i\hbar\delta_{ll'}\partial_{t'}-\varepsilon_{ll'}) +\delta_{ll'}\mathcal{F}_l^{+}(t')\}\varsigma^{-}_{il'}(t-t'), \tag{19}$$

which no longer depends explicitly on the external field; and again for $\mathcal{F}_i^{-}(t)$ analogously. As can be seen from Eq. (19), this equation can be Fourier transformed in frequency, leading to a system of simple algebraic equations

$$\sum_l \Phi^{\bar{\sigma}}_{il}(\omega)\,\frac{\mathcal{F}_l^{\sigma}(\omega)}{\hbar\langle S_l^z\rangle_{\mathrm{eq}}}=\sum_l \mathcal{H}^{\bar{\sigma}}_{il(1)}(\omega)\,\langle S_l^{\sigma}\rangle, \tag{20}$$

where $\Phi^{\sigma}_{il}(\omega)=\langle S_i^z\rangle_{\mathrm{eq}}\,\delta_{il}-\varsigma^{\sigma}_{il}(\omega)$ is the Fourier transform of the generalized function of intensity (17), and

$$\mathcal{H}^{\pm}_{il(1)}(\omega)=\frac{1}{\hbar\langle S^{z}_{l}\rangle_{\mathrm{eq}}}\sum_{l'}\,(\mp\hbar\omega\delta_{ll'}-\varepsilon_{ll'})\varsigma^{\pm}_{il'}(\omega). \tag{21}$$

With the help of linear algebra, we now solve Eq. (20) for $\mathcal{F}^{\sigma}_{i}(\omega)$, leading to an exact expression for the nonequilibrium correlations,

$$\hbar^{-1}\mathcal{F}^{\sigma}_{i}(\omega)=\sum_{l}\,\mathcal{H}^{\bar{\sigma}}_{il}(\omega)\langle S^{\sigma}_{l}(\omega)\rangle. \tag{22}$$

They are expressed in terms of the average of the individual spins and a nonlocal effective magnetic field that contains implicitly all the spin correlations,

$$\mathcal{H}^{\sigma}_{il}(\omega)=\sum_{l'}\,\frac{D^{\sigma}_{il'}(\omega)\langle S^{z}_{i}\rangle_{\mathrm{eq}}}{D^{\sigma}(\omega)}\mathcal{H}^{\sigma}_{l'l(1)}(\omega), \tag{23}$$

where $D^{\sigma}(\omega)=\det||\Phi^{\sigma}_{il}(\omega)||$ is the determinant of the spin-correlation intensity, and $D^{\sigma}_{il}(\omega)$ its algebraic complement. If we would have taken the approximation $\Phi^{\sigma}_{il}(\omega)\approx\langle S^{z}_{i}\rangle_{\mathrm{eq}}\,\delta_{il}$, Eq. (20) would obtain the same form as Eq. (22). Thus the quantity $\mathcal{H}^{\sigma}_{il(1)}$ corresponds to the effective magnetic field (23) to first order in the spin correlations $\varsigma^{\sigma}_{il}(\omega)$. When taking the inverse Fourier transform of Eq. (22) back into the time domain, we obtain the required microscopic analogy of the Landau-Lifshitz equations. Thus, we rewrite Eq. (6) in a form that depends only on the average of an individual spin

$$i\,\partial_{t}\langle S^{+}_{i}\rangle=\gamma[h_{i,\mathrm{eff}}\langle S_{i}\rangle]^{+z}-\sum_{l}\,\int_{-\infty}^{\infty}dt'\,\mathcal{H}^{-}_{il}(t-t')\langle S^{+}_{l}(t')\rangle, \tag{24a}$$

$$-i\,\partial_{t}\langle S^{-}_{i}\rangle=\gamma[h_{i,\mathrm{eff}}\langle S_{i}\rangle]^{-z}-\sum_{l}\,\int_{-\infty}^{\infty}dt'\,\mathcal{H}^{+}_{il}(t-t')\times\langle S^{-}_{l}(t')\rangle, \tag{24b}$$

where the square brackets are now linearized in the external magnetic field,

$$[h_{i,\mathrm{eff}}\langle S_{i}\rangle]^{\sigma z}=\left(h^{\sigma}_{i,\sim}(t)-\frac{1}{g\mu_{B}}\sum_{j\neq i}\,I_{ij}\langle S^{\sigma}_{j}\rangle\right)\langle S^{z}_{i}\rangle_{\mathrm{eq}}-\left(h^{z}_{0}-\frac{1}{g\mu_{B}}\sum_{j\neq i}\,I_{ij}\langle S^{z}_{j}\rangle_{\mathrm{eq}}\right)\langle S^{\sigma}_{i}\rangle,$$

and we made use of the fact that $\langle S^{\pm}\rangle_{\mathrm{eq}}=0$. The time-dependent nonlocal magnetic field is defined as

$$\mathcal{H}^{\sigma}_{il}(t-t')=\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\mathcal{H}^{\sigma}_{il}(\omega)\,e^{-i\omega(t-t')}. \tag{25}$$

The first term on the right-hand side of Eq. (24) couples the cyclic coordinates with the *z* coordinate, i.e., the direction of the spontaneous moment. It is local in time and describes the classical precession of the spin averages coupled indirectly with each other via the Weiss molecular field of the exchange interaction. The second term, however, correlates not only the cyclic coordinates with the *z* coordinate but also the cyclic coordinates among each other. This integral term introduces the effect of "memory" in the system, when the spin oscillations at a given moment depend directly on the oscillations at an earlier time. The time of this memory is given by the interval in $t-t'$ in which the kernel $\mathcal{H}^{\sigma}_{il}(t-t')$ differs significantly from zero. Equations (24) describe then a non-Markovian process.[24]

Further, we would like to stress that with help of Eq. (24) it is straightforward to find and investigate the eigenoscillations of the system which exist for $h^{\sigma}_{i,\sim}(t)=0$. As can be seen from the memory kernel, the properties of these oscillations do not depend on the way they were excited. It would have been much more difficult to show the validity of this assertion if we did not exclude the time-dependent field $h^{\sigma}_{i,\sim}(t')$ from the expression (11). If it is necessary to find the response of the spin system to a magnetic field $h^{\sigma}_{i,\sim}(t)$, Eqs. (24) become nonhomogeneous and their solutions can be written in terms of a superposition of the free oscillations. Equations (24) are closed in the sense that they only include the average of individual spins $\langle S^{\mu}_{i}\rangle$ and in contrary to the equations of motion (6) do not include nonequilibrium functions of higher order.

B. Hierarchy of the linearized equations

Before, it was pointed out that the coherent spin fluctuations, created by the external magnetic field, depend on the thermal and spontaneous fluctuations. As a consequence, we could use linear-response theory to decouple the equation of motion systematically and express the nonequilibrium averages $\langle s^{\mu}_{i}s^{\nu}_{j}\rangle$ in terms of the nonequilibrium averages of the individual spins. All spin correlations of $\mathcal{F}_{i}$ in Eq. (6) are taken care of by the nonlocal effective magnetic field $\mathcal{H}^{\sigma}_{il}$ or the so-called memory kernel which is expressed in terms of the equilibrium three-spin Green's functions (13). Introducing this field allows for a separation of the correlations into a hierarchy: Low-lying spin-wave-type excitations contained in $\mathcal{H}^{\sigma}_{il(1)}$ interact with each other to form excitations of higher order, whose excitation energies are given by the poles of D^{σ}. In other words, the theoretical description can be viewed as a sequence of plateaus, where each plateau is a self-consistent description of the system in analogy to the Landau-theory of Fermi liquids.

For example, the low-lying excitations are described by an ideal magnon gas. The interactions between the magnons lead then to higher-order excitations of the spin system. They correspond either to the scattering of the individual magnons or to their coupling. If the coupled state corresponds to a two-magnon state, Eq. (23) is comparable to what was obtained by Boyd and Callaway on the basis of a "ladder-approximation" in low-temperature perturbation theory.[25] In a similar fashion, one could derive more complex coupled excitations at temperatures close to T_{c}. On the other hand, if the deviations from the equilibrium value of the spin average are small, i.e., $\varsigma^{\sigma}_{il}/\langle S^{z}_{i}\rangle_{\mathrm{eq}}\ll 1$ and $D^{\sigma}(\omega)\neq 0$, so that only the scattering of spin waves has to be taken into account, we can use Eq. (20) to solve for $\mathcal{H}^{\sigma}_{il}$ iteratively instead of applying the exact solution (23). Thus one finds the nth order iteration in the nonlocal magnetic field $\mathcal{H}^{\sigma}_{il(\mathrm{n})}$ to be related to the $(n-1)$th order approximation,

$$\mathcal{H}^{\sigma}_{il(\mathrm{n})}(\omega)=\sum_{l'}\left\{\delta_{il'}\mathcal{H}^{\sigma}_{l'l(1)}(\omega)+\frac{\varsigma^{\sigma}_{il'}(\omega)}{\langle S^{z}_{l}\rangle_{\mathrm{eq}}}\mathcal{H}^{\sigma}_{l'l(\mathrm{n\text{-}1})}(\omega)\right\}, \tag{26}$$

where $\mathcal{H}^{\sigma}_{il(0)}=0$. To first order we obtain again Eq. (21). To second order the solution is equivalent to the result of Dyson's second Born approximation in an infinite bulk system as will be shown further down in the text.[21]

C. Possible approaches to solve the linearized equations

In fact, Eq. (24) is the main result of our paper. The form of the integrodifferential equations have a more universal character than presented here within the Heisenberg model and can be readily generalized to more realistic problems than that of an ideal ferromagnet. In the following, we treat Eq. (24) as an example and show how it can be applied to a large variety of physical systems.

First, there are spin clusters. Since Eq. (24) is a system of $2N$ coupled integrodifferential equations, it can, in principle, be computed to any degree of accuracy, in particular, if the number of spins N in the system is not large. The same holds for the equilibrium value of the spin average, and the three-spin Green's function. This is an interesting problem of its own in computational physics. In such cases one can often neglect the dipole-dipole interactions and directly apply Eq. (24). In a similar fashion, one could treat random spin systems.[26]

A second class presents mesoscopic systems which can be either one- or two-dimensional, or of the multilayer type. Whereas the former can be solved by Fourier transform into momentum space, the latter is no longer translational invariant in the direction perpendicular to the layers in general. However, if the number of layers is not too large, the system of integrodifferential equations (24) can still be solved. The direction, in which the system is translational invariant, can be treated with help of a finite-temperature perturbation theory for spin operators, similar to the one of three-dimensional systems to be discussed later. In addition, one has to adapt the Hamiltonian with regards to the reduced dimensionality of the system, since in two dimensions ferromagnetic order is absent at finite temperatures in the Heisenberg model,[27] in contrast to most of the real systems. For ferromagnetic films either dipole-dipole interactions or anisotropy terms need to be included as discussed by Yafet, Kwo, and Gyorgy[28] or Bander and Mills,[29] respectively.

The last class are macroscopic three-dimensional systems, and we can pursue our analysis of Eq. (24) in the limit where the number of spins is large. The properties of such systems depend only on the relative distance between spins at different sites so that one can take the Fourier transform of Eq. (24) into momentum space. Further, the average $\langle S^{\mu}_{i}\rangle$ is independent of site in equilibrium and is thus relieved of its index, i.e., $\langle S^{z}_{i}\rangle_{\mathrm{eq}}=\langle S^{z}\rangle_{\mathrm{eq}}$. Of course, also for three-dimensional systems the Heisenberg model is only an insufficient description for realistic systems, since, for example, the long-range dipole-dipole forces can never really be neglected due to the physical boundaries of any system. Nevertheless, as pointed out before, the Heisenberg model is a good example to demonstrate the validity of our approach. In particular, to solve Eq. (24) explicitly, we have to find an expression for $\langle S^{z}\rangle_{\mathrm{eq}}$, and the higher spin correlations $\varsigma^{\pm}_{il}$ (12) or the three-spin Green's function $\Gamma^{(r)}$ (13), respectively. These quantities can be found approximatively with help of a finite-temperature perturbation theory for spin operators. In the following, we would like to demonstrate this for the case of an infinite Heisenberg ferromagnet.

IV. DIAGRAMMATIC PERTURBATION THEORY FOR SPIN OPERATORS

There have been various attempts to find an appropriate algorithm for spin-operators in the past. A fundamental contribution was given in particular by the works of Stinchcombe *et al.*[12] and later independently by Vaks, Larkin, and Pikin.[13,14] Whereas Stinchcombe and co-workers used a linked cluster expansion in the spirit of Kahn and Uhlenbeck,[30] Vaks, Larkin, and Pikin introduced fictitious fermion fields. The latter was reformulated by making explicit use of Wick's theorem in an adaptation to spin operators by Izyumov, Kassan-Ogly, and Skryabin.[10,11] Despite the elegance of the formulation, their work seems to have found rather little attention, and we thus take the liberty to restate some parts of it in the following as well as to adapt them to suit the calculation of the three-spin Green's function (13). It turns out that the three-spin Green's function can be described by simple quasiparticle interactions of the spin system.

The problem when dealing with spin operators is based on the fact that they do not commute with each other to yield simple c numbers as can be seen from the relations (2). The algorithms allowing one to calculate the Green's functions for Bose or Fermi particles with simple algebraic relations have to be replaced by a method which is adapted to deal with the consequences of more complicated relations such as $[[[S^{+}_{i},S^{z}_{i}],S^{z}_{i}],S^{-}_{i}]=2\,S^{z}_{i}$. As is already shown with this example, the role of the S^{z} operator is a particular one, since unpaired cyclic operators $S^{\pm}$ vanish, when taking the equilibrium average. The idea is, first, to get rid of all the cyclic operators $S^{\pm}$ in any average expanded to nth order in the interacting part of the Hamiltonian with help of the commutator relations (2), and then to calculate independently the averages of the remaining S^{z} operators. The averages are of the form

$$\langle T\overline{S}^{\mu_i}_{i}(\tau_i)\,\ldots\,\overline{S}^{\mu_l}_{l}(\tau_l)\rangle=\frac{\langle TS^{\mu_i}_{i}(\tau_i)\,\ldots\,S^{\mu_l}_{l}(\tau_l)\mathcal{S}(\beta)\rangle_{(0)}}{\langle\mathcal{S}(\beta)\rangle_{(0)}}, \tag{27}$$

and are written for convenience as thermal or imaginary-time averages, where $\tau=it$, and T orders operators in imaginary time. While dealing with finite-temperature Green's functions, we shall set $\hbar=1$ for convenience. The operators are then given in the imaginary-time Heisenberg representation $\overline{S}^{\mu}(\tau)=\exp(\tau\overline{H})\,S^{\mu}\exp(-\tau\overline{H})$ in the average on the left-hand side, whereas in the averages on the right-hand side by $S^{\mu}(\tau)=\exp(\tau H_0)\,S^{\mu}\exp(-\tau H_0)$. Here, we separated the Hamiltonian into a unperturbed and interacting part. The averages $\langle\ldots\rangle_{(0)}$ in Eq. (27) are thus determined by the density matrix $\rho_0=\exp(-\beta H_0)/\mathrm{Sp}\{\exp(-\beta H_0)\}$ according to Eq. (4), and the finite-temperature S matrix,

$$\mathcal{S}(\beta)=\sum_{n=0}^{\infty}\frac{(-1)^n}{n!}\times\int_0^\beta d\tau_1\ldots\int_0^\beta d\tau_n[TH_{\mathrm{int}}(\tau_1)\ldots H_{\mathrm{int}}(\tau_n)]. \tag{28}$$

Although there are different ways splitting up the equilibrium Hamiltonian in its unperturbed and interacting part, we take for convenience the approach which contains the Weiss molecular field in zeroth-order such that $\bar{H}=(H_0-E_0)+H_{\mathrm{int}}$,

$$E_0=-\tfrac{1}{2}NI_0\langle S^z\rangle_{(0)}^2, \tag{29a}$$

$$H_0=-y\sum_i S_i^z=(g\mu_B h_0^z-I_0\langle S^z\rangle_{(0)})\sum_i S_i^z, \tag{29b}$$

$$H_{\mathrm{int}}=-\tfrac{1}{2}\sum_{i,i'}{}' I_{ii'}(s_i^- s_{i'}^+ + s_i^z s_{i'}^z), \tag{29c}$$

where $I_0=\Sigma_{j\neq i}I_{ij}$. To separate E_0 and H_0 from H_{int}, we once more used a fluctuation operator, $s_i^\mu=S_i^\mu-\langle S^\mu\rangle_{(0)}$, however, this time describing the deviations at equilibrium from the local spin average $\langle S^\mu\rangle_{(0)}$. E_0 is the Weiss energy of the spin system which takes the value $-NI_0S^2/2$ at zero temperature.

Enlarging on the particular role of the S^z operators, we assume for now, that we have transformed the numerator of the right-hand side of Eq. (27) with help of the S matrix (28) and commutator relations (2) into such a form, that we are left with averages containing only S^z operators. It is then possible to calculate these remaining averages from the partition function of an individual spin. This is similar to calculating the magnetization in the Weiss molecular field or self-consistent field approximation. Using the local partition function for an individual spin, i.e., for the constant term E_0 is not accounted:

$$Z=\mathrm{Sp}\exp\left(\beta\sum_i^N y_i S_i^z\right), \tag{30}$$

where the index at y is introduced for convenience, we obtain for the averages over S^z operators

$$\langle S_1^z\ldots S_n^z\rangle_{(0)}=\frac{\partial_{y_1}\ldots\partial_{y_n}Z}{\beta^n Z}. \tag{31}$$

A single-operator average is then proportional to the Brillouin function $B_S(x)=[1+(2S)^{-1}]\,\mathrm{cth}[(S+1/2)x]-(2S)^{-1}\,\mathrm{cth}(x/2)$ as in the Weiss molecular field theory, $\langle S_1^z\rangle_{(0)}=SB_S(\beta y)=b(\beta y)$, whereas a n-operator average contains the nth-order derivative of the Brillouin function, e.g., $\langle S_1^zS_2^z\rangle_{(0)}=b^2+b'\,\delta_{12}$. The first term on the right-hand side of the last example is the product of two spin averages and the second term accounts for the interactions in the molecular field approximation. In other words, a method is obtained that reproduces in its zeroth order the molecular field result beyond which it allows to calculate, at least in principle, all the contributions to any thermodynamic variable.

There is one important point to stress. To make contact with the theories of Stinchcombe and co-workers[12] as well as Vaks, Larkin, and Pikin[13,14] we included the Weiss molecular field in the unperturbed Hamiltonian. This assumes that the exchange interaction has a large radius of interaction, so that it is possible to expand in powers of H_{int}.

A. Wick's theorem for spin operators

Before discussing the details of Wick's theorem, we would like to give an example and calculate the following average:

$$\langle TS_2^+(\tau_2)S_1^-(\tau_1)\rangle_{(0)}=\begin{cases}\langle S_2^+(\tau_2)S_1^-(\tau_1)\rangle_{(0)} & \text{if} \quad \tau_1<\tau_2\\ \langle S_1^-(\tau_1)S_2^+(\tau_2)\rangle_{(0)} & \text{if} \quad \tau_1>\tau_2.\end{cases} \tag{32}$$

Since the time development of the spin operators has the usual form, $S^+(\tau)=\exp(-y\tau)S^+$ and $S^-(\tau)=\exp(y\tau)S^-$, we obtain with the help of Eq. (2),

$$\langle S_2^+(\tau_2)S_1^-(\tau_1)\rangle_{(0)}=\langle S_1^-(\tau_1)S_2^+(\tau_2)\rangle_{(0)}+2\,\delta_{12}\,e^{-y(\tau_2-\tau_1)}\langle S_2^z\rangle_{(0)}. \tag{33}$$

This can be transformed, by using the invariance of the trace under cyclic permutations and $S^+(\tau)\rho_0=\exp(-\beta y)\,\rho_0\,S^+(\tau)$, into

$$\langle S_2^+(\tau_2)S_1^-(\tau_1)\rangle_{(0)}=2\,\delta_{12}\,e^{-y(\tau_2-\tau_1)}(n_y+1)\langle S_2^z\rangle_{(0)}, \tag{34}$$

where $n_y=\{\exp(\beta y)-1\}^{-1}$ is the distribution function for Bose particles. We have thus reduced the average of two spin operators to an average over a single S^z operator. It is now possible to define an unperturbed time-ordered Green's function,

$$K_{12}^{(0)}(\tau_1-\tau_2)=-\frac{1}{2}\frac{\langle TS_2^+(\tau_2)S_1^-(\tau_1)\rangle_{(0)}}{\langle S_2^z\rangle_{(0)}}=-\delta_{12}\,e^{-y(\tau_2-\tau_1)}\begin{cases}n_y+1 & \text{if} \quad \tau_1<\tau_2\\ n_y & \text{if} \quad \tau_1>\tau_2.\end{cases} \tag{35}$$

This Green's function is not yet the spin-wave Green's function of the true one-spin-wave, or one-magnon eigenstates. To obtain the proper one-magnon Green's function, we have to sum up an infinite series in the perturbation expansion as will be shown later. However, the necessary diagram rules have to be introduced first.

The Wick's theorem for spin operators states that all possible pairings between S^- and S^+ as well as between S^- and S^z have to be taken under the condition that they should be time ordered.[10,11] This means that we replace in the initial average $\langle TS_i^{\mu_i}(\tau_i)\ldots S_{n'}^{\mu_{n'}}(\tau_{n'})\rangle_{(0)}$, as given by the numerator on the right-hand side of Eq. (27), a pair of operators with one of the relations

$$S_1^-(\tau_1)S_2^z(\tau_2)=+K_{12}^{(0)}(\tau_1-\tau_2)\,S_2^-(\tau_2), \tag{36a}$$

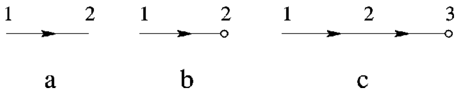

FIG. 1. Diagrams according to the pairing relations (36).

$$S_1^-(\tau_1)S_2^+(\tau_2)=-2\,K_{12}^{(0)}(\tau_1-\tau_2)\,S_2^z(\tau_2). \tag{36b}$$

In this way site ‘‘1’’ cannot take part in further pairings. In Eq. (36a) the operator S_2^- needs to be paired up again, however site ‘‘2’’ can take part only in one further pairing which results in the relation

$$S_1^-(\tau_1)S_2^z(\tau_2)S_3^+(\tau_3) \\ =-2\,K_{12}^{(0)}(\tau_1-\tau_2)K_{23}^{(0)}(\tau_2-\tau_3)\,S_3^z(\tau_3). \tag{36c}$$

There is still one more possible pairing. The operator in Eq. (36b) S_2^z can be left as it stands or used for pairing up another S^- operator. This changes the operator at site ‘‘2’’ into another S^- operator which needs to paired up again, so that site ‘‘2’’ takes part in three and only three pairings. This can be expressed in a way as illustrated in the following example:

$$S_1^-(\tau_1)S_2^+(\tau_2)S_3^-(\tau_3)S_4^+(\tau_4) \\ =2^2K_{12}^{(0)}(\tau_1-\tau_2)K_{23}^{(0)}(\tau_2-\tau_3)K_{34}^{(0)}(\tau_3-\tau_4)\,S_4^z(\tau_4). \tag{36d}$$

Thus, the initial average can be reduced to a product of Green’s functions $K^{(0)}$, averages of the S^z operators, which can in turn be calculated via relation (31), and a numerical factor of $(-2)^{m_+}$, where m_+ is the number of pairings of type (36b). A convenient way to represent the pairing relations (36) is in the form of diagrams as shown in Fig. 1, where $K^{(0)}$ is presented by a full line starting from an S^- operator, and the m remaining S^z operators are drawn as a circles.

B. Rules for constructing diagrams

We would like to calculate an average containing l spin operators. An expansion to the order n given by

$$\langle TS_i^{\mu_i}(\tau_i)\ldots S_l^{\mu_l}(\tau_l)S_1^{\mu_1}(\tau_1)S_{1'}^{\mu_1'}(\tau_{1'})\ldots \\ \times S_n^{\mu_n}(\tau_n)S_{n'}^{\mu_n'}(\tau_{n'})\rangle_{(0)}$$

similar to that on the right-hand side of Eq. (27) contains n operators H_{int}. Each operator H_{int} depends on a pair of sites i and i' over which one has to sum. After we have made use of the rules (36) to draw all possible pairings, the exchange interaction between these pairs $I_{ii'}$ is represented graphically as a wavy line to connect all the $2n$ internal operators with each other. The essential difference to standard perturbation theory is now in calculating the averages over the m remaining S^z operators. They form so-called blocks according to relation (31), where a δ function connecting two sites corresponds graphically to a dashed line between two circles denoting the remaining S^z operators.

The denominator on the right-hand side of Eq. (27) contains only internal lines. Therefore, the corresponding diagrams, the vacuum polarization graphs, cancel exactly all those diagrams in the numerator that are not fully connected with an external operator either through the exchange interaction or by forming blocks. Although this does not imply that all external operators have to be connected with each other, we replace for $l>1$ all S^z-operators by their fluctuation part s^z in the initial average to describe only the correlation effects, e.g., $\langle s_1^z s_2^z\rangle_{(0)}=b'\,\delta_{12}$. In order to calculate the nth-order expansion to an average containing l external operators, one therefore has to draw only all connected topological nonequivalent diagrams, where each diagram corresponds to an analytical expression of the following kind:

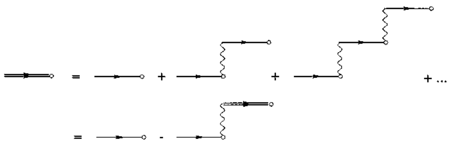

FIG. 2. Molecular field approximation for K_{-+}.

$$(-2)^{m_+}\frac{\mathcal{P}_n}{2^n n!}\prod_m b^{[m-1]} \\ \times\int_0^\beta d\tau_1\ldots\int_0^\beta d\tau_n\sum_{1,1'}{}' I_{11'}\ldots \\ \times\sum_{n,n'}{}' I_{nn'}K^{(0)}\ldots K^{(0)}\delta\ldots\delta. \tag{37}$$

The factor $\mathcal{P}_n$ is the number of topological equivalent diagrams, and $b^{[m-1]}$ is the $(m-1)$th derivative of b.[10,11]

C. Derivation of the one-magnon Green’s function

As pointed out before, the Green’s function $K^{(0)}$ is not compatible with the one-magnon eigenfunction. To obtain the one-magnon Green’s function, we calculate an infinite series of diagrams as shown in Fig. 2. This series is of the Dyson form, and, applying the diagram rules (37), we derive the dressed Green’s function

$$b^{-1}K_{-+}(i,j;\tau-\tau')=K_{ij}^{(0)}(\tau-\tau')-\sum_{1,1'}{}' I_{11'} \\ \times\int_0^\beta d\tau_1\,K_{i1'}^{(0)}(\tau-\tau_1) \\ \times K_{-+}(1,j;\tau_1-\tau'). \tag{38}$$

In Eq. (38) a negative sign occurs due to the fact that the operator s_1^- is internal for $K_{-+}(i,j;\tau-\tau')$ but external for $K_{-+}(1,j;\tau_1-\tau')$, whereas the overall sign in front of a diagram is only determined by the number m_+ of internal operators. Taking the complex conjugate and the Fourier transform of Eq. (38) in time and space, we obtain

$$K_{+-}(\mathbf{k};\omega_n)=\int_0^\beta d\tilde{\tau}\,e^{i\omega_n\tilde{\tau}}\sum_{\mathbf{r}_{ij}} K_{+-}(i,j;\tilde{\tau})e^{-i\mathbf{k}\mathbf{r}_{ij}} \\ =\frac{b}{i\omega_n-\varepsilon_{\mathbf{k}}}, \tag{39}$$

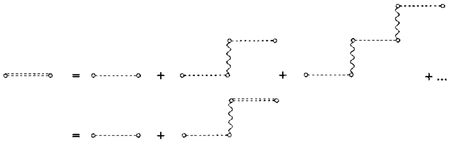

FIG. 3. Molecular field approximation for K_{zz}.

where $\tilde{\tau}=\tau-\tau'$, $\mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j$, $\omega_n=2\pi n/(\beta)$, and $\varepsilon_{\mathbf{k}}=-g\mu_B h_0^z+b(I_0-I_{\mathbf{k}})$ are indeed the one-magnon eigenenergies. In a similar fashion the correlation between the z components of two spins can be calculated as shown in Fig. 3:

$$K_{zz}(\mathbf{k};\omega_n)=\frac{b'}{\beta^{-1}-b'I_{\mathbf{k}}}\delta_{n0}, \tag{40}$$

which is static and cannot couple to a time-dependent magnetic field. Close to zero temperature its contribution is exponentially small when $b^{[n]}\propto\exp(-\gamma_n T_c/T)$. The result of Eq. (39) was obtained independently by Tyablikov,[31] Larkin,[32] and Englert,[33] whereas Eq. (40) was obtained by de Gennes and Villain,[34] as well as Brout and Englert.[35] We need both of them in the following.

D. Calculation of the spin correlations

Now, we evaluate diagrammatically the three-spin Green's function $\Gamma_{z+,-}(i,j;l;\tau-\tau')=-(1/2)\times\langle Ts_i^z(\tau)s_j^+(\tau)s_l^-(\tau')\rangle$, whose Fourier transform shall be defined as

$$\Gamma_{z+,-}(i,j;l;\tilde{\tau}) =\frac{1}{\beta}\sum_n e^{-i\omega_n\tilde{\tau}}\frac{1}{N^2}\sum_{\mathbf{k},\mathbf{q}} e^{i[\mathbf{k}\mathbf{r}_l+(\mathbf{q}-\mathbf{k})\mathbf{r}_i-\mathbf{q}\mathbf{r}_j]}\,\Gamma_{z+,-}(\mathbf{q},\mathbf{k};\omega_n). \tag{41}$$

Its perturbation expansion is represented in Fig. 4. Here, we did not include second-order diagrams that form closed loops in the exchange interaction, since their leading term is of a higher order in $H_{\rm int}$.

The first diagram in the series is made of three parts: a straight line connecting s_l^- with s_i^z, a curved line connecting s_i^z with s_j^+, and a circle representing the remaining s^z operator. Thus, the diagram corresponds to pairing relation (36c). Drawing the diagram in a slightly different manner to the one shown in Fig. 1(c), should draw attention to the fact that the operators s_i^z and s_j^+ are at equal times. Therefore, the corresponding analytical expression in momentum and energy space is

$$-b\,K_{\mathbf{k}}^{(0)}(-\omega_n)K_{\mathbf{q}}^{(0)}(0)=-\frac{b\,n_y}{i\omega_n-y}, \tag{42}$$

where we used the convention that $\tau'<\tau$. This result is independent of $\mathbf{k}$ and $\mathbf{q}$ since we used only the local Green's function $K^{(0)}$ and not the magnon Green's function K_{+-}. In order to obtain the self-consistent solution for $\Gamma_{z+,-}$, it

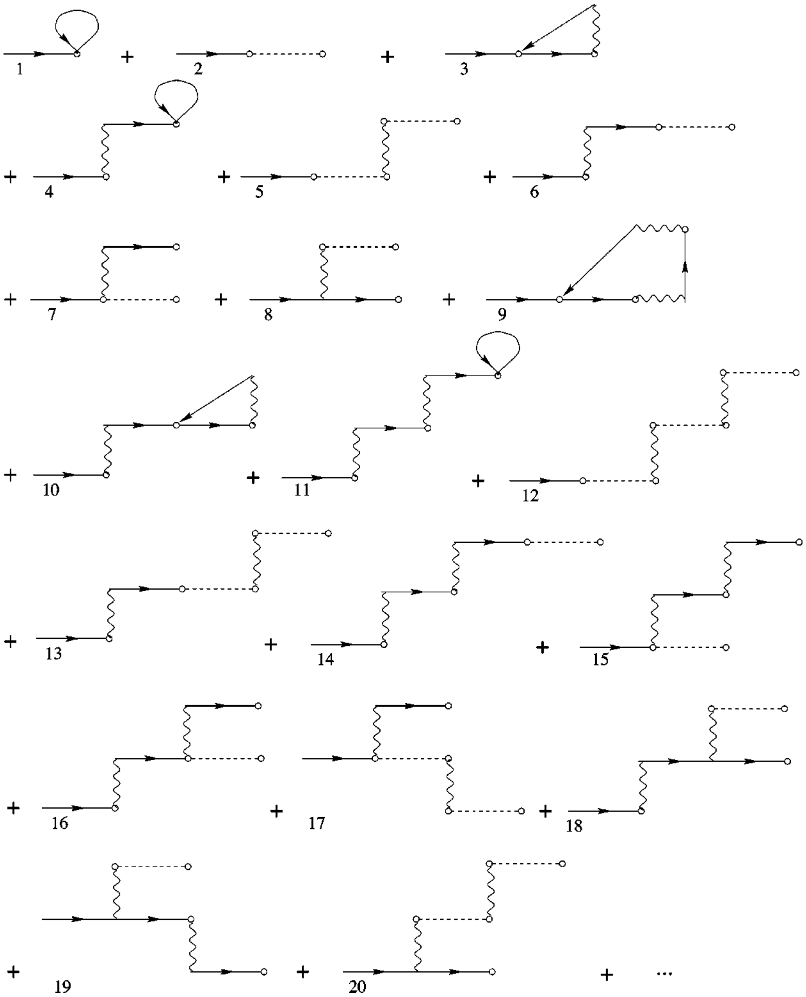

FIG. 4. Diagrams for $\Gamma^{(r)}_{z-,+}$, or $\Gamma^{(r)}_{z+,-}$. When $\omega_n\leftrightarrow-\omega_n$.

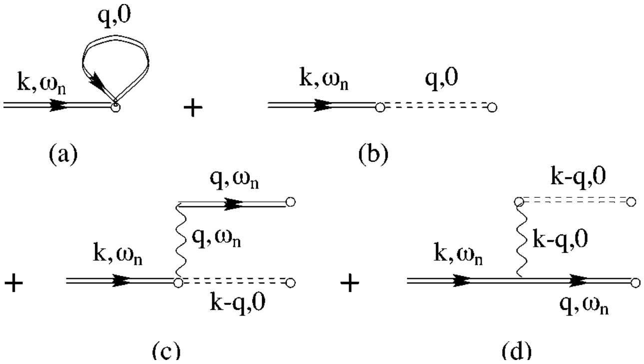

FIG. 5. Dressed diagrams of self-consistent solution for Γ.

therefore seems natural to replace $K^{(0)}$ with K_{+-} given in Eq. (39). That this is indeed possible can be seen from Fig. 4. Besides diagram 1 we have to consider the diagrams 3, 4, 9, 10, and 11 which can be divided into three types. The first type is a series of diagrams only within the pairing between the operators s_l^- and s_i^z and is represented by the diagrams 4 and 11. The second type is a series of diagrams (3 and 9) within the operators at equal times. Finally, diagram 10 contains a mixture of the previous two types. This taken together, yields a product of two infinite sums of diagrams, which can be drawn as shown in Fig. 5(a),

$$\Gamma^{\mathrm{a}}_{z+,-}(\mathbf{q},\mathbf{k};\omega_n)=-\,b^{-1}K_{+-}(\mathbf{k};\omega_n)K_{+-}(\mathbf{q})=-\,\frac{b\,n_{\mathbf{q}}}{i\omega_n-\varepsilon_{\mathbf{k}}}, \tag{43a}$$

where $K_{+-}(\mathbf{q})$ contains the frequency summation implicitly so that it gives the number of spin waves in the molecular field approximation, $n_{\mathbf{q}}=\{\exp(\beta\varepsilon_{\mathbf{q}})-1\}^{-1}$. Turning to the second diagram in Fig. 4, we note that the pairing is only between the operators s_l^- and s_j^+ according to rule (36b). This yields another s^z operator at site j which forms a block with s_i^z. By interchanging the sites i and j for the pairing, the Fourier transform in momentum space differs by the factor $\exp(i(\mathbf{k}-2\mathbf{q})\mathbf{r}_{ij})$ from the one in Eq. (41). Instead, we could equally well interchange the indices z and $+$ in the definition for $\Gamma_{z+,-}$. In other words, the Fourier transform of $\Gamma_{z+,-}$ or $\Gamma_{+z,-}$ does not correspond to its inverse transform, only to that of their linear combination. On the other hand, $\Gamma_{z+,-}-\Gamma_{+z,-}$ is just the quantity which is needed in Eq. (24). However, in order to keep the writing at minimum the exponential factor $\exp(i(\mathbf{k}-2\mathbf{q})\mathbf{r}_{ij})$ is introduced as a dummy variable instead. As for the first diagram in Fig. 4 there is also for this second diagram a subseries of diagrams (6, 14 ; 5, 12 ; 13), which also consists of a product of two infinite sums, and can be dealt with in a similar fashion to yield diagram 5(b):

$$\Gamma^{\mathrm{b}}_{z+,-}(\mathbf{q},\mathbf{k};\omega_n)=b^{-1}K_{+-}(\mathbf{k};\omega_n)K_{zz}(\mathbf{q})e^{i(\mathbf{k}-2\mathbf{q})\mathbf{r}_{ij}}=\frac{e^{i(\mathbf{k}-2\mathbf{q})\mathbf{r}_{ij}}}{i\omega_n-\varepsilon_{\mathbf{k}}}\,\frac{b'}{1-\beta b' I_{\mathbf{q}}}. \tag{43b}$$

There are still two more subseries of diagrams shown in Fig. 4 which contribute to the self-consistent solution of $\Gamma_{z+,-}$. One of the subseries contains the diagrams 7, 15, 16, and 17, while the other one contains 8, 18, 19, and 20. Each of the diagrams has three extensions instead of two and, therefore, the corresponding subseries is a product of three infinite sums as shown in Figs. 5(c) and 5(d),

$$\begin{aligned}\Gamma^{\mathrm{c}}_{z+,-}(\mathbf{q},\mathbf{k};\omega_n)&=-b^{-2}K_{+-}(\mathbf{k};\omega_n)e^{i(\mathbf{k}-2\mathbf{q})\mathbf{r}_{ij}}\\&\quad\times K_{+-}(\mathbf{q};\omega_n)\,I_{\mathbf{q}}K_{zz}(\mathbf{k}-\mathbf{q})\\&=-\frac{e^{i(\mathbf{k}-2\mathbf{q})\mathbf{r}_{ij}}}{i\omega_n-\varepsilon_{\mathbf{k}}}\,\frac{1}{i\omega_n-\varepsilon_{\mathbf{q}}}\,\frac{b' I_{\mathbf{q}}}{1-\beta b' I_{\mathbf{k}-\mathbf{q}}},\end{aligned} \tag{43c}$$

$$\begin{aligned}\Gamma^{\mathrm{d}}_{z+,-}(\mathbf{q},\mathbf{k};\omega_n)&=-b^{-2}K_{+-}(\mathbf{k};\omega_n)K_{+-}(\mathbf{q};\omega_n)\\&\quad\times I_{\mathbf{k}-\mathbf{q}}K_{zz}(\mathbf{k}-\mathbf{q})\\&=-\frac{1}{i\omega_n-\varepsilon_{\mathbf{k}}}\,\frac{1}{i\omega_n-\varepsilon_{\mathbf{q}}}\,\frac{b' I_{\mathbf{k}-\mathbf{q}}}{1-\beta b' I_{\mathbf{k}-\mathbf{q}}}.\end{aligned} \tag{43d}$$

In Eq. (43c) the minus sign occurs for the same reason as in Eq. (39). Since one of the infinite sums was always taken at equal times, $\Gamma^{\mathrm{a}}_{z+,-}$ Eq. (43a) and $\Gamma^{\mathrm{b}}_{z+,-}$ Eq. (43b) essentially take only two-spin correlations into account. The spin waves are weighted with regards to other spin waves Fig. 5(a) or fluctuating spin moments Fig. 5(b), as if they would travel in an ''effective medium.'' On the other hand, $\Gamma^{\mathrm{c}}_{z+,-}$ and $\Gamma^{\mathrm{d}}_{z+,-}$ describe true three-spin correlations. In particular, they correspond to the scattering of spin waves at fluctuating spin moments. There are two ways this can happen; either a spin wave is absorbed by the spin moment which excites a new spin wave with different momentum as shown in Fig. 5(c), or a spin wave is scattered by transferring part of its momentum to the spin moment as follows from Fig. 5(d).

The retarded Green's function can now be obtained by analytical continuation $i\omega_n\rightarrow\omega+i\delta$. We also have to change the sum over frequencies $\beta^{-1}\Sigma_n$ into an integral over frequency $(2\pi)^{-1}\int d\omega$. Performing the integration, we find

$$\Gamma^{(r)}_{z+,-}(\mathbf{q},\mathbf{k};t-t')=K^{(r)}_{+-}(\mathbf{k};t-t')\,\gamma^{(r)}_{z+,-}(\mathbf{q},\mathbf{k};t-t'), \tag{44}$$

where we split off the magnon Green's function from its modulating part

$$K^{(r)}_{+-}(\mathbf{k};t-t')=-\frac{ib}{\hbar}\,\Theta(t-t')\,e^{-i\omega_{\mathbf{k}}(t-t')}, \tag{45a}$$

$$\begin{aligned}\gamma^{(r)}_{z+,-}(\mathbf{q},\mathbf{k};t-t')&=-n_{\mathbf{q}}+\frac{b'}{b}\,\frac{e^{i(\mathbf{k}-2\mathbf{q})\mathbf{r}_{ij}}}{1-\beta b' I_{\mathbf{q}}}\\&\quad-\frac{b'}{b}\,\frac{1-e^{-(i/\hbar)b(I_{\mathbf{k}}-I_{\mathbf{q}})(t-t')}}{1-\beta b' I_{\mathbf{k}-\mathbf{q}}}\\&\quad\times\frac{I_{\mathbf{q}}e^{i(\mathbf{k}-2\mathbf{q})\mathbf{r}_{ij}}+I_{\mathbf{k}-\mathbf{q}}}{I_{\mathbf{k}}-I_{\mathbf{q}}},\end{aligned} \tag{45b}$$

where $\omega_{\mathbf{k}}=\varepsilon_{\mathbf{k}}/\hbar$. It was established earlier in the text that the Green's function $\Gamma^{(r)}_{+z,-}$ is equivalent to $\Gamma^{(r)}_{z+,-}$ when interchanging the sites i and j. Thus, one is left with only the

Green's function $\Gamma^{(r)}_{z-,+}$, since it can be transformed into $\Gamma^{(r)}_{-z,+}$ by the analogous operation. However, the relation $\Gamma^{(r)}_{z-,+}=(\Gamma^{(r)}_{z+,-})^{\dagger}$ found in Sec. III saves us from analyzing $\Gamma^{(r)}_{z-,+}$ separately. Finally, we are in a position to write down an expression for the spin correlations (12)

$$\varsigma^{\bar{\sigma}}_{il}(t-t')=\sum_{j\neq i} I_{ij}\frac{1}{N^2}\sum_{\mathbf{k,q}} (e^{i\mathbf{qr}_{ij}}e^{i\mathbf{kr}_{li}} - e^{-i\mathbf{qr}_{ij}}e^{i\mathbf{kr}_{lj}})\Gamma^{(r)}_{z\sigma,\bar{\sigma}}(\mathbf{q,k};t-t'). \tag{46}$$

V. RESULTING FORM OF THE LINEARIZED EQUATIONS OF MOTION FOR AN INFINITE HEISENBERG FERROMAGNET

With help of Eq. (46), we now calculate the nonlocal effective magnetic field (21), whose Fourier transform reads in the molecular-field approximation as,

$$\mathcal{H}^{-}_{il(1)}(t-t')=-\frac{1}{\hbar b}\sum_{l'} (i\hbar\,\delta_{ll'}\partial_{t'}+\varepsilon_{ll'})\varsigma^{-}_{il'}(t-t'). \tag{47}$$

For clarity, we try to avoid writing both cyclic indices, since the other component can always be obtained taking the complex conjugate. In Eq. (47) the operator is transformed into momentum space such that $\varepsilon_{ll'}$ is substituted by $\varepsilon_{\mathbf{k}}$. We operate on expression (46) and derive

$$\mathcal{H}^{-}_{il(1)}(t-t')=\hbar^{-1}\sum_{j\neq i} I_{ij}\frac{1}{N^2}\sum_{\mathbf{k,q}} (e^{i\mathbf{qr}_{ij}}e^{i\mathbf{kr}_{li}}-e^{-i\mathbf{qr}_{ij}}e^{i\mathbf{kr}_{lj}}) \times[\delta(t-t')-i\hbar\, b^{-1}K^{(r)}_{+-}(\mathbf{k};t-t') \times\partial_{t'}]\gamma^{(r)}_{z+,-}(\mathbf{q,k};t-t'). \tag{48}$$

Examining the square brackets, it follows that it is possible to split up this expression into a part which is local in time and one which is nonlocal in time,

$$\overline{\mathcal{H}}^{-}_{il(1)}(t-t')=-\hbar^{-1}\sum_{j\neq i} I_{ij}R_{ij}(T)(\delta_{li}-\delta_{lj})\delta(t-t'), \tag{49a}$$

$$\tilde{\mathcal{H}}^{-}_{il(1)}(t-t')=\frac{b'}{b\hbar}\sum_{j\neq i} I_{ij}\frac{1}{N^2}\sum_{\mathbf{k,q}}\frac{I_{\mathbf{k-q}}-I_{\mathbf{q}}}{1-\beta b' I_{\mathbf{k-q}}} \times(e^{i\mathbf{qr}_{ij}}e^{i\mathbf{kr}_{li}}-e^{-i\mathbf{qr}_{ij}}e^{i\mathbf{kr}_{lj}})K^{(r)}_{+-}(\mathbf{q};t-t'), \tag{49b}$$

respectively. The first two terms in Eq. (45b) could be absorbed into a temperature-dependent correlation parameter between two spins, describing the effective spin medium

$$R_{ij}(T)=N^{-1}\sum_{\mathbf{q}} e^{i\mathbf{qr}_{ij}}\left(n_{\mathbf{q}}+\frac{b'}{b}\frac{1}{1-\beta b' I_{\mathbf{q}}}\right). \tag{50}$$

The third term in Eq. (45b) gave rise to expression (49b). As the summation over **k** and **q** can no longer be factorized, the spins at sites *i*, *j*, and *l* are fully correlated with each other. The underlying physical process is the scattering of spin waves from the fluctuating *z* component of a spin as discussed earlier in the text and shown in Fig. 5(c) and 5(d).

Before we can write down the resulting form of Eq. (24), there is still a quantity left which is not yet calculated beyond the molecular field result. This is the average of the spontaneous moment at equilibrium $\langle S^z\rangle_{\text{eq}}$. As this is a well-known result, we just state it in the following and refer to Ref. 13 for further details:

$$\langle S^z\rangle_{\text{eq}(1)}=\frac{1}{N}\sum_{\mathbf{q}}\left((n_y-n_{\mathbf{q}})+b'\beta I_{\mathbf{q}}n_{\mathbf{q}}+\frac{b''}{2}\frac{\beta I_{\mathbf{q}}}{1-\beta b' I_{\mathbf{q}}}\right). \tag{51}$$

The first term is the spin-wave correction which gives the characteristic Bloch law $(T/T_c)^{3/2}$ for $\langle S^z\rangle_{\text{eq}}$ at low temperature. The second term gives a correction from the scattering of spin waves at the spontaneous moment, whereas the last term describes the effect of fluctuations of the spontaneous moment on itself.

Inserting the effective fields of the two-spin correlations (49a) and of the three-spin correlations (49b) into Eq. (24), we obtain the resulting form of the linearized equations of motion for individual spin averages

$$\pm i\,\gamma^{-1}\partial_t\langle S_i^{\pm}\rangle=h^{\pm}_{i,\sim}\langle S^z\rangle_{\text{eq}}-h^z_0\langle S_i^{\pm}\rangle+\frac{\langle S^z\rangle_{\text{eq}}}{g\mu_B}\sum_{j\neq i} I^{\text{eff}}_{ij}(T) \times(\langle S_i^{\pm}\rangle-\langle S_j^{\pm}\rangle)-\sum_{l}\int_{-\infty}^{\infty}dt'\,\tilde{\mathcal{H}}^{\mp}_{il(1)} \times(t-t')\langle S_l^{\pm}(t')\rangle. \tag{52}$$

With help of the correlation parameter (50), we defined a temperature-dependent effective exchange interaction $I^{\text{eff}}_{ij}(T)=I_{ij}(1+R_{ij}(T)/\langle S^z\rangle_{\text{eq}})$. This gives manifestation to the empirical fact that the exchange interaction should, in principle, depend on temperature.[23]

So far, the terms in the perturbation series were considered according to their expansion in powers of H_{int}. On the other hand, the method allows us to classify diagrams with respect to the derivatives of *b*, in other words, to their temperature behavior. In particular, at low temperatures derivatives of *b* can be neglected due to their exponential smallness.

Taking thus the low-temperature approximation of Eq. (52), the last term, nonlocal in time, drops out. The equations of motion for an individual spin average is then rather concise. The average spin moment (51) takes the form $\langle S^z\rangle_{\text{eq}}\approx S-R(T)$, where $R(T)=N^{-1}\Sigma_{\mathbf{q}}n_{\mathbf{q}}$. The spin-correlation parameter depends also no longer on b', i.e. $R_{ij}(T)=N^{-1}\Sigma_{\mathbf{q}}n_{\mathbf{q}}\exp(i\mathbf{qr}_{ij})$. This low-temperature form of Eq. (52) corresponds to the results of Ref. 9 and justifies the approximations taken therein.

In the remaining part of this section we would like to answer the questions of how our approach corresponds to previous results on the thermodynamics of the three-dimensional Heisenberg ferromagnet, and how to obtain from our results macroscopic equations of the magnetization which include temperature dependence and damping of the spin excitations.

A. Spectrum and Landau damping

To demonstrate how to obtain the spectrum from Eq. (52), we deal with its last term first. We assume that the spin waves are monochromatic, i.e., $\langle S_i^+\rangle \propto \exp(-i\omega t)$. The time integration yields

$$\int_{-\infty}^{\infty} dt'\, K_{+-}^{(r)}(\mathbf{q};t-t')e^{-i\omega t'} = \frac{b}{\hbar}\left\{\wp\left(\frac{1}{\omega-\omega_{\mathbf{q}}}\right)-i\pi\,\delta(\omega-\omega_{\mathbf{q}})\right\}e^{-i\omega_{\mathbf{q}}t}, \tag{53}$$

where $\wp$ denotes the principle part. The first term contributes to the spectrum. The second term gives characteristic Landau damping of the spin waves. Looking for a solution of Eq. (52) in an infinite crystal, so that the spin waves depend periodically on $\mathbf{r}_i$, i.e., $s_i^+=N^{-1}\Sigma_{\mathbf{k}}s^+(\mathbf{k})\exp(i\mathbf{k}\mathbf{r}_i)$, we find for the spectrum and damping constant

$$E_{\mathbf{k}}=\varepsilon_{\mathbf{k}}-N^{-1}\sum_{\mathbf{q}}\left\{(I_{\mathbf{k}-\mathbf{q}}-I_{\mathbf{q}})n_{\mathbf{q}}+\frac{b'}{b}\frac{I_{\mathbf{k}-\mathbf{q}}-I_{\mathbf{q}}}{1-\beta b' I_{\mathbf{q}}} - \frac{b'}{\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{q}}}\frac{(I_{\mathbf{k}-\mathbf{q}}-I_{\mathbf{q}})^2}{1-\beta b' I_{\mathbf{k}-\mathbf{q}}}\right\}, \tag{54a}$$

$$\gamma_{\mathbf{k}}=\frac{b'\pi}{\hbar N}\sum_{\mathbf{q}}\frac{(I_{\mathbf{k}-\mathbf{q}}-I_{\mathbf{q}})^2}{1-\beta b' I_{\mathbf{k}-\mathbf{q}}}\,\delta(\omega-\omega_{\mathbf{q}}). \tag{54b}$$

Vaks, Larkin, and Pikin reported this result first.[14] Comparing Eq. (54a) with Eq. (51), it is evident that the temperature dependence of the spin-wave spectrum corresponds only to zeroth order with that of $\langle S^z\rangle_{\text{eq}}$. Due to the exponential smallness of b' at low temperatures, damping is only noticeable for intermediate temperatures and rises sufficiently together with the fluctuations as the temperature gets in the vicinity of T_c. However, for very long spin waves the damping is still weak even close to the critical region. We do not expand the discussion of this point and just refer to the cited references for further details, since the purpose of this section was to show the validity of our approach by making contact to previous works in the field.

B. Macroscopic equation for the magnetization

In the following, we shall take the classical limit of the low temperature approximation of Eq. (52), to derive the macroscopic magnetization equations at low-temperatures. These equations will contain the previous convention, that $\mathbf{h}_0$ is in the negative z direction and $\mathbf{h}_\sim(t)$ is perpendicular to $\mathbf{h}_0$.

The macroscopic magnetization is defined as, $\mathbf{m}(\mathbf{r})=-g\mu_B\langle\mathbf{S}(\mathbf{r})\rangle V_0^{-1}$, where $\langle\mathbf{S}(\mathbf{r})\rangle|_{\mathbf{r}=\mathbf{r}_i}=\langle\mathbf{S}_i\rangle$ and V_0 the volume of the unit cell, and the exchange integral as, $J(\mathbf{r}-\mathbf{r}')|_{\mathbf{r}=\mathbf{r}_i,\mathbf{r}'=\mathbf{r}_j}=I_{ij}V_0$. Since the exchange integral is falling off rapidly with increasing $|\mathbf{r}-\mathbf{r}'|$, the magnetization can be developed by a Taylor expansion in powers of $\mathbf{s}=\mathbf{r}-\mathbf{r}'$, where $\mathbf{s}$ is a vector along the crystallographic axes. Taking the series expansion to second order, we make the following transformation:

$$\frac{1}{g\mu_B}\sum_{j\neq i} I_{ij}^{\text{eff}}(T)(\langle S_i^{\pm}\rangle-\langle S_j^{\pm}\rangle) = \frac{1}{2(g\mu_B)^2}\sum_{\mathbf{s}} J^{\text{eff}}(\mathbf{s},T)\,(\mathbf{s}\cdot\boldsymbol{\nabla})^2 m^{\pm}(\mathbf{r},T)\equiv h_{\text{ex}}^{\pm}(\mathbf{r},T), \tag{55}$$

where $J^{\text{eff}}(\mathbf{s},T)=J(\mathbf{s})\{1+R(\mathbf{s},T)/|\mathbf{m}_0(T)|\}$, and $\mathbf{m}_0(T)$ is the macroscopic equilibrium magnetization. The sum over $\mathbf{s}$ is usually performed only to include nearest- or next-to-nearest-neighbor coupling. In correspondence to the magnetization density, the two-spin correlation parameter is taken at the long-wavelength approximation $\lambda \gg a$:

$$R(\mathbf{s},T)=-\,g\mu_B\int_{-\infty}^{\infty}\frac{dq^3}{2(2\pi)^3}\,\frac{2-(\mathbf{s}\cdot\mathbf{q})^2}{e^{\beta\varepsilon_{\mathbf{q}}}-1}, \tag{56}$$

where $\varepsilon_{\mathbf{q}}=g\mu_B|\mathbf{h}_0|+(S/2)\Sigma_{\mathbf{s}}J(\mathbf{s})\,(\mathbf{s}\cdot\mathbf{q})^2$, and V_0 is the volume of the unit cell. The parameter $R(T)$ differs from the one defined above, in that the second term in the numerator of definition (56) is absent.

Apart from the temperature dependence of the exchange integral, the expression on the right-hand side of Eq. (55) is analogous to the effective field contribution derived from the ‘‘classical exchange energy’’ in the Landau and Lifshitz equations of motion. Therefore, we defined it as a temperature-dependent effective exchange field $h_{\text{ex}}^{\pm}(\mathbf{r},T)$. Using Cartesian coordinates, the equations of motion (52) are generalized towards the following form in their classical limit at low temperatures:

$$\gamma^{-1}\partial_t\,\mathbf{m}(\mathbf{r},T)=\mathbf{h}_0\times\mathbf{m}(\mathbf{r},T)+[\mathbf{h}_\sim(\mathbf{r})+\mathbf{h}_{\text{ex}}(\mathbf{r},T)]\times\mathbf{m}_0(T), \tag{57}$$

where we dropped again for brevity the argument denoting the time dependence. The equations of motion (57) are the temperature-dependent equivalent of the linearized Landau-Lifshitz equations of motion without damping. The temperature dependence of $J^{\text{eff}}(\mathbf{s},T)$ cancels the spurious $T^{3/2}$ effect in the spin-wave spectrum as required. In other words, the contributions that arise in the equations of motion (57) due to the correlation of thermal fluctuations $R(\mathbf{s},T)$, are of the same order as those of the spontaneous magnetization $\mathbf{m}_0(T)$. Equation (57) can be seen as the principle extension of the Landau-Lifshitz equations of motion to macroscopic systems at low temperatures, where $J^{\text{eff}}(\mathbf{s},T)$ and $\mathbf{m}_0(T)$ can be obtained, either from first principles as shown, or taken as phenomenological constants from experiment. Moreover, the equations of motion (57) allow for solving boundary problems, since in the long-wavelength limit $\lambda \gg a$, we can usually assume $\lambda \gg a\sqrt{T_c/T}$ so that the physical surface of the system does not influence the correlation parameter and equilibrium magnetization.

In order to include damping at low temperatures in the equations of motion (57), we have to consider spin-wave-spin-wave interactions.[21] One way would be to calculate Eq. (52) to next order, applying the equation-of-motion technique. This would lead to a four-spin Green's function which could be calculated to first order at low temperatures by just replacing $K^{(0)}$ with K_{+-} and neglecting terms containing b' in the perturbation expansion. Finally, the limit of long wavelengths has to be taken. The correction to the molecular field approximation would then be of second order in H_{int}. Another more straightforward method was introduced in Sec. III B. Iterating the nonlocal field $\overline{\mathcal{H}}^{\sigma}_{il(1)}$ Eq. (49a) with the help of Eqs. (26) and (46), one obtains for the nonequilibrium correlations at low temperatures $\mathcal{F}^{\pm}_{i}=\mathcal{F}^{\pm}_{i(1)}+\mathcal{F}^{\pm}_{i(2)}$, where

$$\mathcal{F}^{+}_{i(1)}(t)=\sum_{j\neq i} I_{ij}R_{ij}[\langle S^{+}_{j}(t)\rangle-\langle S^{+}_{i}(t)\rangle], \tag{58a}$$

$$\begin{aligned}\mathcal{F}^{+}_{i(2)}(t)=b^{-1}\sum_{j\neq i} I_{ij}R_{ij}{\sum_{l,l'}}' I_{ll'}R_{ll'}\int_{-\infty}^{\infty}dt'[K^{(r)}_{+-}(i,l;t-t')\\ -K^{(r)}_{+-}(j,l;t-t')][\langle S^{+}_{l}(t')\rangle-\langle S^{+}_{l'}(t')\rangle],\end{aligned} \tag{58b}$$

and $\mathcal{F}^{-}_{i(1)}$, $\mathcal{F}^{-}_{i(2)}$ by taking the complex conjugate. The Fourier transform of the magnon Green's function reads $K^{(r)}_{+-}(i,j;t-t')=N^{-1}\Sigma_{\mathbf{k}}K^{(r)}_{+-}(\mathbf{k};t-t')\exp i\mathbf{k}\mathbf{r}_{ij}$. The term $\mathcal{F}^{\pm}_{i(1)}$ features already in Eqs. (52) and (57), respectively. The term $\mathcal{F}^{\pm}_{i(2)}$ is new and with Eq. (53) can be seen to give damping. It corresponds to the results of Dyson's second Born approximation.[21] Without doubt, within the presented formalism it is rather straightforward to derive this result, since no additional Green's functions need to be calculated.

In order to include also $\mathcal{F}^{\pm}_{i(2)}$ in the macroscopic equations of motion (57), we first express the magnon Green's function in terms of x and y coordinates,

$$\begin{aligned}K^{(r)}_{xx}(i,j;t-t')&=\frac{1}{2}[K^{(r)}_{+-}(i,j;t-t')+K^{(a)}_{+-}(i,j;t-t')]\\ &=K^{(r)}_{yy}(i,j;t-t'),\end{aligned} \tag{59a}$$

$$\begin{aligned}K^{(r)}_{yx}(i,j;t-t')&=\frac{1}{2i}[K^{(r)}_{+-}(i,j;t-t')-K^{(a)}_{+-}(i,j;t-t')]\\ &=-K^{(r)}_{xy}(i,j;t-t').\end{aligned} \tag{59b}$$

This allows us to define a 3×3 matrix $\mathbf{K}^{(r)}=|K^{(r)}_{\alpha\beta}|$, where $\alpha,\beta=x,y,z$ and $K^{(r)}_{z\alpha}=K^{(r)}_{\alpha z}=0$, so that the corresponding vector to the nonequilibrium correlations (58b) takes in Cartesian coordinates the form

$$\begin{aligned}\boldsymbol{\mathcal{F}}_{i(2)}(t)=b^{-1}\sum_{j\neq i} I_{ij}R_{ij}{\sum_{l,l'}}' I_{ll'}R_{ll'}\int_{-\infty}^{\infty}dt'[\mathbf{K}^{(r)}(i,l;t-t')\\ -\mathbf{K}^{(r)}(j,l;t-t')][\langle \mathbf{S}_{l}(t')\rangle-\langle \mathbf{S}_{l'}(t')\rangle].\end{aligned} \tag{60}$$

When taking the long-wavelength limit of $b^{-1}K^{(r)}_{+-}(i,j;t-t')$, we derive the following function:

$$\begin{aligned}K^{(r)}_{+-}(\mathbf{s};t-t')=-\frac{iV_0}{\hbar}\Theta(t-t')\int_{-\infty}^{\infty}\frac{dk^3}{2(2\pi)^3}\\ \times[2-(\mathbf{k}\cdot\mathbf{s})^2]e^{-(i/\hbar)\varepsilon_{\mathbf{k}}(t-t')},\end{aligned} \tag{61}$$

where $\mathbf{s}=\mathbf{r}_i-\mathbf{r}_j$. We also dropped the linear term in $\mathbf{k}$ since it cancels, when performing the integral. Now, we are in the position to write the nonequilibrium correlations $\boldsymbol{\mathcal{F}}_{i(2)}(t)$ in the macroscopic limit

$$\begin{aligned}\boldsymbol{\mathcal{F}}_{(2)}(\mathbf{r},t)=\sum_{\mathbf{s},\mathbf{s}',\mathbf{s}''}\int_{-\infty}^{\infty}dt'\ \mathbf{R}(\mathbf{s},\mathbf{s}',\mathbf{s}'',t-t')\\ \times\mathbf{h}_{\mathrm{ex}}(\mathbf{s}',\mathbf{r}-\mathbf{s}'',t'),\end{aligned} \tag{62}$$

where the kernel

$$\begin{aligned}\mathbf{R}(\mathbf{s},\mathbf{s}',\mathbf{s}'',t-t')=-(g\mu_B)^{-1}J(\mathbf{s})R(\mathbf{s},T)R(\mathbf{s}',T)\\ \times[\mathbf{K}^{(r)}(\mathbf{s}'',t-t')-\mathbf{K}^{(r)}(\mathbf{s}''-\mathbf{s},t-t')]\end{aligned}$$

contains all information on the correlations. The matrix $\mathbf{K}^{(r)}(\mathbf{s},t-t')$ is the long-wavelength limit of $b^{-1}\mathbf{K}^{(r)}(i,j;t-t')$. The quantity $\mathbf{h}_{\mathrm{ex}}(\mathbf{r},\mathbf{s})=[1/2(g\mu_B)^2]J(\mathbf{s})\,(\mathbf{s}\cdot\boldsymbol{\nabla})^2\mathbf{m}(\mathbf{r},T)$, is defined similar as before and $\mathbf{h}_{\mathrm{ex}}(\mathbf{r})=\Sigma_{\mathbf{s}}\mathbf{h}_{\mathrm{ex}}(\mathbf{r},\mathbf{s})$. In order to include the term (62) in the equations of motion (57), we substitute $\mathcal{F}^{x}_{(2)}\rightarrow-\mathcal{F}^{y}_{(2)}$ and $\mathcal{F}^{y}_{(2)}\rightarrow\mathcal{F}^{x}_{(2)}$. This new correlation term is responsible for what is known in classical spin-wave theory as a four-magnon scattering process, which is hinted at by the four different space arguments in the sum of the term (62).

VI. CONCLUSIONS

The standard Landau-Lifshitz equations are extensively used in magnetism due to the important fact that they allow one to solve boundary problems in a rather straightforward manner. They provide a method to calculate magnetic oscillations and waves in finite magnetic samples. The new equations (24) and (57) obtained in this work generalize the linearized Landau-Lifshitz equations in two important aspects: First, the equations (24) are microscopic and are therefore applicable to ferromagnetic systems of any type of dimensionality and geometry. Second, Eqs. (24) and (57) include the influence of spin correlations and not only the effect of an effective molecular field. The presence of spin correlations leads to the possibility to describe the temperature dependence of the dynamic properties in the underlying magnetic system. In addition, Eqs. (24) and (57) are equally suitable and convenient as the Landau-Lifshitz equations to solve boundary problems. In fact, the structure of the macroscopic equations (57) does not differ, in principle, from that of the Landau-Lifshitz equations. Only a temperature dependence of the individual coefficients occurs. As concerns the microscopic equations (24), they turn into a system of ordinary differential equations, after Fourier transformation in time.

Nevertheless, there still remains one principle problem. It is, of course, necessary to know the individual coefficients and their temperature dependence in the differential equations (24) and (57). However, the same problem exists in the case of the Landau-Lifshitz equations, where one needs to know the constants for exchange, anisotropy etc. As a rule, they are taken from phenomenological considerations, i.e., from the requirement to obtain agreement between theory and experiment. This can obviously be done also in our case. However, there is in addition the possibility to derive the necessary coefficients from first principles. These coefficients contain the information on the equilibrium properties of the system, and in the present work we obtained for them expressions in terms of equilibrium averages of spin operators, i.e., three-spin Green's functions as given in Eqs. (12) and (13).

In order to find the appropriate algorithm to calculate these spin averages, we discussed different classes of systems in Sec. III C. In Sec. IV we gave a demonstration for a particular example, that of an infinite Heisenberg ferromagnet. Since the thermodynamic properties of such a simple system are already studied in detail, we were able to compare our results with established ones obtained by different methods.[12–14] This gave the possibility of demonstrating the validity of our approach and its relative simplicity.

In particular, at the heart of our approach lies the combination of different analytical tools. The equation-of-motion technique for spin operators is employed to derive the principle equations of motion (6) and to single out the spin correlations (9). We used linear-response theory to decouple the equations of motion and express the nonequilibrium averages $\langle s_i^\mu s_j^\nu \rangle$ in terms of the nonequilibrium averages of the individual spins. The obtained closed equations (19) contain the nonequilibrium spin correlations to all orders given the equilibrium properties of the system are known. As a result, the decoupling was obtained in a systematic manner and free of any arbitrariness, which might arise in the equation of motion technique, when applied to Green's functions for spin operators. It is interesting to pursue this approach to higher orders, to obtain closed equations for clusters of spins.

The next step of our analysis was to calculate the equilibrium properties diagrammatically. Besides the spontaneous magnetization, we needed to find the three-spin Green's function (13). At first sight, one would expect that the analysis of the three-spin Green's functions is very involved, but it turned out that indeed the opposite is the case. Namely, to obtain the first-order correction to the molecular field approximation, one has to sum-up diagrams that form simple combinations of geometric progressions. This led to the intuitive picture, that the underlying physical processes are simple quasiparticle interactions of the spin system. In order to obtain the spin-wave spectrum, we could have inserted the expression for the spin correlations (46) directly into Eq. (15) and evaluated the poles of the two-spin Green's function in this approximation, instead of solving the equations of motion (52). In other words, by combining the equation-of-motion technique with diagrammatic perturbation theory, an effective method is obtained to derive higher-order corrections to the molecular field result. One simply has to calculate higher-order Green's functions self-consistently and then insert them into the corresponding equation of motion. The method keeps, thus, a simplicity which lacks in ordinary diagrammatic perturbation theory, due to the rapidly increasing number of different types of diagrams for each higher order in the expansion. Not only the number of diagrams which have to be analyzed is significantly reduced, but also the underlying physical process seems presented more lucidly. We look forward to see this method employed also in other areas of condensed-matter physics.

The use of the simple Heisenberg model and the comparison to well-known results can be regarded as a prerequisite for the future development of research in this direction. We plan to use our approach to solve actual boundary problems. For instance, we are interested in the temperature behavior of macroscopic spin waves in ferrite films of the YIG type. As such films are used in electronic devices, one outstanding problem is how to obtain films whose spectral characteristics are most stable against temperature fluctuations. A further direction we currently focus on was already mentioned in the context of small magnetic clusters and two-dimensional magnetic nanostructures of the multilayer type. Last but not least, another problem we are working on, is to drop the assumption of linear response in order to study the influence of correlations on nonlinear and parametric effects.

ACKNOWLEDGMENTS

We would like to thank academician Yu. V. Gulyaev, Professor Sir R. J. Elliott, and Professor R. B. Stinchcombe for their interest in our work. P.E.Z. would like to acknowledge the Russian Foundation of Basic Research, and C.H. acknowledges the support provided by the HSK III of the German State and Länder via the DAAD.

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