Some effects of kinematical interactions in a ferromagnet

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The Heisenberg model of a ferromagnet is analyzed by a semiclassical method that attempts to include the effects of both dynamical and kinematical interactions. At low temperatures, the results of Dyson are reproduced. At temperatures where the kinematical interactions become important, a gap appears in the magnon excitation spectrum. This gap results, in part, from a long-range three-magnon effective interaction. The interaction consists of the renormalization of the self-energy of a magnon on some lattice site whenever an appreciable number of sites elsewhere in the crystal contain two or more magnon wave packets. Because of the long range, the presence of a gap does not violate the Goldstone theorem. An expression is derived for the magnon excitation energy which includes the effects of n-magnon effective interactions.

I. INTRODUCTION

Let us consider the Heisenberg model[1] of a three-dimensional ferromagnet. The one-magnon Bloch spin-wave states[2] are orthonormal exact excited-state eigenfunctions of the Heisenberg Hamiltonian. In contrast, the multiple-magnon spin-wave states have the defects of being (a) not exact eigenfunctions of the Hamiltonian, and (b) neither normalized nor orthogonal. In his definitive treatment of the subject, Dyson[3] used property (a) to define the dynamical interaction and property (b) to define the kinematical interaction. In physical terms, the dynamical interaction refers to the attractive interaction between neighboring, localized magnon wave packets; the kinematical interaction refers to the ability of one-magnon wave packet to inhibit or (in the spin-$\frac{1}{2}$ case) prevent a second magnon wave packet's occupancy of the same lattice site. Dyson developed a power-series expansion of the free energy of the system, an expansion in ascending powers of temperature. Let us write

$$F=F_0+\Delta F\ , \tag{1}$$

where F_0 is the free energy in the absence of any kinematical interactions, and ΔF is the net contribution to the free energy resulting from kinematical interactions. Dyson proved the remarkable results that the power-series expansion of ΔF vanishes identically. This does not mean that ΔF itself vanishes. Although it must vanish at $T=0$, it certainly does not vanish at temperatures close to the Curie temperature T_C. The presumption is that ΔF is negligible at temperatures small compared with T_C.

We make the corresponding decomposition for the internal energy

$$U=U_0+\Delta U\ , \tag{2}$$

where ΔU is the net contribution to the internal energy resulting from kinematical interactions. Let us try to understand qualitatively the nature of ΔU. As already mentioned, the spin-wave states are not properly normalized when there are two or more magnons in the entire crystal. At low temperatures the effects of this must be negligible. We assume that the lack of normalization becomes important when an appreciable fraction of all the lattice sites of the crystal are occupied by two or more magnons. We let n_0 be the mean number of magnons per lattice site. We consider the probability that a given site contains two or more magnons. By expanding this probability as a power series in n_0, the leading term will be proportional to n_0^2. Similarly, the leading term in the expansion of the renormalization factor for the magnon spin waves will also be proportional to n_0^2. In turn, there will be a term proportional to n_0^2 in the renormalization of the magnon excitation energies. Thus there will be a contribution to ΔU of the form

$$\Delta U_1 \equiv c_1 n_0^2 \sum_k \hbar\omega_{k0} f_k\ . \tag{3}$$

Here c_1 is a constant, $\hbar\omega_{k0}$ is the excitation energy of a magnon of wave vector $\vec{k}$ in the absence of kinematical interactions, and f_k is the statistical Bose factor. We write

$$\hbar\omega_k=\hbar\omega_{k0}+\Delta_k\ , \tag{4}$$

where Δ_k is the net contribution to $\hbar\omega_k$ resulting from kinematical interactions. Note that ΔU_1 is an effective *three-body interaction.* It is also an effective *long-range interaction,* in that the magnon being renormalized can be far removed from the magnons giving rise to the renormalization. We have

$$\hbar\omega_k=(\partial U/\partial f_k)\ , \tag{5}$$

$$n_0=N^{-1}\sum_k f_k\ , \tag{6}$$

N being the total number of lattice sites in the crystal. It follows that there is a contribution to Δ_k (and thus to $\hbar\omega_k$) of the form

$$\Delta_{k1}\equiv(\partial/\partial f_k)\Delta U_1=c_1 n_0^2\left[\hbar\omega_{k0}+(n_0N)^{-1}\sum_{k'} 2f_{k'}\hbar\omega_{k'0}\right]. \tag{7}$$

Note that in the $k=0$ limit where $\hbar\omega_{k0}$ vanishes, Δ_{k1} stays finite, with the value

$$\Delta_{01}=c_1 n_0 N^{-1}\sum_{k'} 2f_{k'}\hbar\omega_{k'0} . \quad (8)$$

We have arrived at the remarkable conclusion that there is a *gap* in the magnon excitation spectrum, by virtue of the kinematical interaction. Furthermore, there is no conflict with the strictures of the Goldstone theorem,[4] since the effective interaction is long range. In order to be consistent with Dyson's results for ΔF, the quantity Δ_k should have a vanishing power-series expansion in T. For example, if Δ_k varies as $T^{-n}\exp[-(T_C/T)]$, this condition results. It could well be that this behavior results from the presence of a gap in the excitation spectrum. It should be admitted that the present arguments ignore the question of whether or not a magnon excitation energy can even be defined, because of lifetime effects, at temperatures close to T_C.

In the next section, we will develop a method, admittedly approximate, that attempts to deal with both dynamical and kinematical interactions. A key ingredient of the method is the use of the so-called semiclassical approximation,[5] which allows for the treatment of finite angles of spin deviation without getting into the morass of nonphysical sectors of Hilbert space. The resulting expressions for the magnon excitation energy can be decomposed into a series of terms associated with the various types of effective multiple-magnon interactions. Through two-body interactions, the results coincide with Dyson's results at low temperatures. A gap in the excitation spectrum first arises with three-body interactions and has a form qualitatively consistent with Eq. (8).

II. SEMICLASSICAL TREATMENT

The Heisenberg Hamiltonian is

$$H=-\tfrac{1}{2}\sum_{i,j} J_{ij}\vec{S}_i\cdot\vec{S}_j , \quad (9)$$

$$J_{ij}=J(|\vec{R}_i-\vec{R}_j|),\ \ J_{ii}=0 , \quad (10)$$

where the double sum is over the N lattice sites of the crystal. Making use of the commutation properties of angular momentum,

$$\vec{S}_i\times\vec{S}_j=i\delta_{ij}\vec{S}_i , \quad (11)$$

we can write

$$d\vec{S}_i/dt=\vec{S}_i\times\vec{H}_i , \quad (12)$$

$$\vec{H}_i\equiv-\hbar^{-1}\delta H/\delta\vec{S}_i=\hbar^{-1}\sum_j J_{ij}\vec{S}_j . \quad (13)$$

Equation (12) is the correct quantum-mechanical equation of motion for $d\vec{S}_i/dt$. Invoking the semiclassical approximation,[5] we try to solve the equation classically. One would expect the approximation to be especially good in treating the magnon excitation energy in the long-wavelength limit $\vec{k}=\vec{0}$, where the total spin of the crystal is precessing as a unit.[6]

We take the system in the absence of any magnons to be in that state where all spins are pointing along the x axis, i.e., an orientation given by the spherical coordinates $\theta_0=\frac{1}{2}\pi$, $\phi_0=0$. We choose this orientation in order to avoid certain technical problems associated with the z direction (problems related to the fact that the spherical coordinate ϕ_0 is undefined in that direction). In the presence of an assembly of magnons, the spin on the ith lattice site will point in the direction $\theta_0+\theta_i,\phi_0+\phi_i$, where

$$\begin{aligned}\phi_i&=\sum_k C_k\sin(\vec{k}\cdot\vec{R}_i-\omega_k t+\Phi_k) ,\\ \theta_i&=\sum_k C_k\cos(\vec{k}\cdot\vec{R}_i-\omega_k t+\Phi_k) .\end{aligned} \quad (14)$$

Note that ϕ_i and θ_i each represent rotational oscillations with respect to axes perpendicular to the ground-state orientation. Together, ϕ_i and θ_i represent a series of precessions about the equilibrium orientation. Φ_k is a random-phase angle associated with the mode $\vec{k}$. C_k is the amplitude of the mode $\vec{k}$. In the interest of simplicity, we restrict ourselves to the spin-$\frac{1}{2}$ case. The number of magnons of wave vector $\vec{k}$ associated with the amplitude C_k is given by

$$f_k\equiv\tfrac{1}{2}N(1-\cos C_k)=\tfrac{1}{4}NC_k^2 ,$$

so that

$$C_k^2=4N^{-1}f_k . \quad (15)$$

Thus C_k is proportional to $N^{-1/2}$ and is very small. However, ϕ_i and θ_i are not necessarily small. At any instant of time, the spin on the ith lattice site will have an angular velocity perpendicular to the direction of the spin.

We wish to perform a sequence of two oscillatory transformations of coordinates, a different set for each lattice site, given by

$$\begin{aligned}S_{ix1}&\equiv S_{ix}\cos\phi_i+S_{iy}\sin\phi_i ,\\ S_{iy1}&\equiv -S_{ix}\sin\phi_i+S_{iy}\cos\phi_i ,\\ S_{iz1}&\equiv S_{iz} ,\\ S_{ix2}&\equiv S_{ix1}\cos\theta_i-S_{iz1}\sin\theta_i ,\\ S_{iy2}&\equiv S_{iy1} ,\\ S_{iz2}&\equiv S_{ix1}\sin\theta_i+S_{iz1}\cos\theta_i .\end{aligned} \quad (16)$$

The idea is to make the precessing spins appear static after the two transformations, the ith spin pointing along the ith x_2 axis. Interchanging the order of the transformations, we have

$$\begin{aligned}S_{ix1}&\equiv S_{ix}\cos\theta_i-S_{iz}\sin\theta_i ,\\ S_{iy1}&\equiv S_{iy} ,\\ S_{iz1}&\equiv S_{ix}\sin\theta_i+S_{iz}\cos\theta_i ,\\ S_{ix2}&\equiv S_{ix1}\cos\phi_i+S_{iy1}\sin\phi_i ,\\ S_{iy2}&\equiv -S_{ix1}\sin\phi_i+S_{iy1}\cos\phi_i ,\\ S_{iz2}&\equiv S_{iz1} .\end{aligned} \quad (17)$$

Since the two transformations do not commute, these two sequences will lead to different results. As we will see, it is necessary to take a particular linear combination of the results of the two sequences. Because of the noncommutivity of the two transformations, it follows that for either of the two sequences there will be an instantaneous component of angular velocity *parallel* to the instantaneous orientation of the spin on a given lattice site. This spurious parallel component of angular velocity is associated with the first of the two transformations in either sequence. In order to mitigate the effects of the spurious component, we will follow the two oscillatory transformations by a rotational transformation parallel to the x_2 axis, the same for all lattice sites. Thus we take

$$\begin{aligned} S_{ix3} &\equiv S_{ix2} , \\ S_{iy3} &\equiv S_{iy2}\cos(\Omega t) - S_{iz2}\sin(\Omega t) , \\ S_{iz3} &\equiv S_{iy2}\sin(\Omega t) + S_{iz2}\cos(\Omega t) . \end{aligned} \tag{18}$$

We will later choose an explicit value for Ω.

We first consider sequence I [Eq. (16)] of the two oscillatory transformations. We have

$$\begin{aligned} \frac{dS_{ix1}}{dt} &= \dot{\phi}_i S_{iy1} + \frac{dS_{ix}}{dt}\cos\phi_i + \frac{dS_{iy}}{dt}\sin\phi_i , \\ \frac{dS_{iy1}}{dt} &= -\dot{\phi}_i S_{ix1} - \frac{dS_{ix}}{dt}\sin\phi_i + \frac{dS_{iy}}{dt}\cos\phi_i , \\ \frac{dS_{iz1}}{dt} &= \frac{dS_{iz}}{dt} . \end{aligned} \tag{19}$$

We define $H_{ix1}, H_{iy1}, H_{iz1}$ such that

$$\begin{aligned} \frac{dS_{ix1}}{dt} &= S_{iy1}H_{iz1} - S_{iz1}H_{iy1} , \\ \frac{dS_{iy1}}{dt} &= S_{iz1}H_{ix1} - S_{ix1}H_{iz1} , \\ \frac{dS_{iz1}}{dt} &= S_{ix1}H_{iy1} - S_{iy1}H_{ix1} , \end{aligned} \tag{20}$$

Thus

$$\begin{aligned} H_{ix1} &= H_{ix}\cos\phi_i + H_{iy}\sin\phi_i \\ &= \hbar^{-1}\sum_j J_{ij}[\cos(\phi_j - \phi_i)S_{jx1} - \sin(\phi_j - \phi_i)S_{jy1}] , \\ H_{iy1} &= -H_{ix}\sin\phi_i + H_{iy}\cos\phi_i \\ &= \hbar^{-1}\sum_j J_{ij}[\sin(\phi_j - \phi_i)S_{jx1} + \cos(\phi_j - \phi_i)S_{jy1}] , \\ H_{iz1} &= \dot{\phi}_i + H_{iz} = \dot{\phi}_i + \hbar^{-1}\sum_j J_{ij}S_{jz1} . \end{aligned} \tag{21}$$

We have

$$\begin{aligned} \frac{dS_{ix2}}{dt} &= -\dot{\theta}_i S_{iz2} + \frac{dS_{ix1}}{dt}\cos\theta_i - \frac{dS_{iz1}}{dt}\sin\theta_i , \\ \frac{dS_{iy2}}{dt} &= \frac{dS_{iy1}}{dt} , \\ \frac{dS_{iz2}}{dt} &= +\dot{\theta}_i S_{ix2} + \frac{dS_{ix1}}{dt}\sin\theta_i + \frac{dS_{iz}}{dt}\cos\theta_i . \end{aligned} \tag{22}$$

The corresponding values of $H_{ix2}, H_{iy2}, H_{iz2}$ are

$$\begin{aligned} H_{ix2} &= H_{ix1}\cos\theta_i - H_{iz1}\sin\theta_i , \\ H_{iy2} &= \dot{\theta}_i + H_{iy1} , \\ H_{iz2} &= H_{ix1}\sin\theta_i + H_{iz1}\cos\theta_i . \end{aligned} \tag{23}$$

In particular,

$$\begin{aligned} H_{ix2} = -\dot{\phi}_i\sin\theta_i + \hbar^{-1}\sum_j J_{ij}\{&[\cos(\phi_j - \phi_i)\cos\theta_i\cos\theta_j + \sin\theta_i\sin\theta_j]S_{jx2} \\ &- \sin(\phi_j - \phi_i)\cos\theta_i S_{jy2} + [\cos(\phi_j - \phi_i)\cos\theta_i\sin\theta_j - \sin\theta_i\cos\theta_j]S_{jz2}\} , \end{aligned} \tag{24}$$

$$H_{iy2} = \dot{\theta}_i + \hbar^{-1}\sum_j J_{ij}[\sin(\phi_j - \phi_i)(\cos\theta_j S_{jx2} + \sin\theta_j S_{jz2}) + \cos(\phi_j - \phi_i)S_{jy2}] , \tag{25}$$

$$\begin{aligned} H_{iz2} = \dot{\phi}_i\cos\theta_i + \hbar^{-1}\sum_j J_{ij}\{&[\cos(\phi_j - \phi_i)\sin\theta_i\cos\theta_j - \cos\theta_i\sin\theta_j]S_{jx2} \\ &- \sin(\phi_j - \phi_i)\sin\theta_i S_{jy2} + [\cos(\phi_j - \phi_i)\sin\theta_i\sin\theta_j + \cos\theta_i\cos\theta_j]S_{jz2}\} . \end{aligned} \tag{26}$$

We next consider sequence II [Eq. (17)] of the two oscillatory transformations. In a similar fashion, we find

$$\begin{aligned} H_{ix2} = \dot{\theta}_i\sin\phi_i + \hbar^{-1}\sum_j J_{ij}\{&[\cos(\theta_j - \theta_i)\cos\phi_i\cos\phi_j + \sin\phi_i\sin\phi_j]S_{jx2} \\ &+ [-\cos(\theta_j - \theta_i)\cos\phi_i\sin\theta_j + \sin\phi_i\cos\phi_j]S_{jy2} + \sin(\theta_j - \theta_i)\cos\phi_i S_{jz2}\} , \end{aligned} \tag{27}$$

$$\begin{aligned} H_{iy2} = \dot{\theta}_i\cos\theta_i + \hbar^{-1}\sum_j J_{ij}\{&[-\cos(\theta_j - \theta_i)\sin\phi_i\cos\phi_j + \cos\phi_i\sin\phi_j]S_{jx2} \\ &+ [\cos(\theta_j - \theta_i)\sin\phi_i\sin\phi_j + \cos\phi_i\cos\phi_j]S_{jy2} - \sin(\theta_j - \theta_i)\sin\phi_i S_{jz2}\} , \end{aligned} \tag{28}$$

$$H_{iz2}=\dot{\phi}_i+\hbar^{-1}\sum_j J_{ij}[-\sin(\theta_j-\theta_i)(\cos\phi_j S_{jx2}-\sin\phi_j S_{jy2})+\cos(\theta_j-\theta_i)S_{jz2}] \ . \tag{29}$$

Clearly, the results obtained for H_{ix2},H_{iy2},H_{iz2} depend on the order in which the transformations are performed. We choose a normalized linear combination of the two sequences, i.e.,

$$H_{iu2}=\tfrac{1}{2}(1+\epsilon)H_{iu2}(\mathrm{I})+\tfrac{1}{2}(1-\epsilon)H_{iu2}(\mathrm{II}), \quad u=x,y,z \ . \tag{30}$$

For the time being we leave the parameter ϵ arbitrary. Note that the case $\epsilon=0$ corresponds to choosing the symmetrized product of the two oscillatory transformations.

We introduce the notation

$$H_{iu2}=A_{iu}+\hbar^{-1}\sum_{j,v} J_{ij}B_{ijuv}S_{jv2}, \quad u,v=x,y,z \ . \tag{31}$$

Thus

$$A_{ix}=-\tfrac{1}{2}[(1+\epsilon)\dot{\phi}_i\sin\theta_i-(1-\epsilon)\dot{\theta}_i\sin\phi_i] \ , \tag{32}$$

$$A_{iy}=\tfrac{1}{2}\dot{\theta}_i[(1+\epsilon)+(1-\epsilon)\cos\phi_i] \ , \tag{33}$$

$$A_{iz}=\tfrac{1}{2}\dot{\phi}_i[(1+\epsilon)\cos\theta_i+(1-\epsilon)] \ , \tag{34}$$

$$B_{ijxx}=\tfrac{1}{2}(1+\epsilon)[\cos(\phi_j-\phi_i)\cos\theta_i\cos\theta_j+\sin\theta_i\sin\theta_j]+\tfrac{1}{2}(1-\epsilon)[\cos(\theta_j-\theta_i)\cos\phi_i\cos\phi_j+\sin\phi_i\sin\phi_j] \ , \tag{35}$$

$$B_{ijxy}=-\tfrac{1}{2}(1+\epsilon)\sin(\phi_j-\phi_i)\cos\theta_i-\tfrac{1}{2}(1-\epsilon)[\cos(\theta_j-\theta_k)\cos\phi_i\sin\phi_i-\sin\phi_i\cos\phi_j] \ , \tag{36}$$

$$B_{ijxz}=\tfrac{1}{2}(1+\epsilon)[\cos(\phi_j-\phi_i)\cos\theta_i\sin\theta_j-\sin\theta_i\cos\theta_j]+\tfrac{1}{2}(1-\epsilon)\sin(\theta_j-\theta_i)\cos\phi_i \ , \tag{37}$$

$$B_{ijyx}=\tfrac{1}{2}(1+\epsilon)\sin(\phi_j-\phi_i)\cos\theta_j+\tfrac{1}{2}(1-\epsilon)[-\cos(\theta_j-\theta_i)\sin\phi_i\cos\phi_j+\cos\phi_i\sin\phi_j] \ , \tag{38}$$

$$B_{ijyy}=\tfrac{1}{2}(1+\epsilon)\cos(\phi_j-\phi_i)+\tfrac{1}{2}(1-\epsilon)[\cos(\theta_j-\theta_i)\sin\phi_i\sin\phi_j+\cos\phi_i\cos\phi_j] \ , \tag{39}$$

$$B_{ijyz}=\tfrac{1}{2}(1+\epsilon)\sin(\phi_j-\phi_i)\sin\theta_j-\tfrac{1}{2}(1-\epsilon)\sin(\theta_j-\theta_i)\sin\phi_i \ , \tag{40}$$

$$B_{ijzx}=\tfrac{1}{2}(1+\epsilon)[\cos(\phi_j-\phi_i)\sin\theta_i\cos\theta_j-\cos\theta_i\sin\theta_j]-\tfrac{1}{2}(1-\epsilon)\sin(\theta_j-\theta_i)\cos\phi_j \ , \tag{41}$$

$$B_{ijzy}=-\tfrac{1}{2}(1+\epsilon)\sin(\phi_j-\phi_i)\sin\theta_i+\tfrac{1}{2}(1-\epsilon)\sin(\theta_j-\theta_i)\sin\phi_j \ , \tag{42}$$

$$B_{ijzz}=\tfrac{1}{2}(1+\epsilon)[\cos(\phi_j-\phi_i)\sin\theta_i\sin\theta_j+\cos\theta_i\cos\theta_j]+\tfrac{1}{2}(1-\epsilon)\cos(\theta_j-\theta_i) \ . \tag{43}$$

We now introduce the essential approximation of *linearizing* the A's with respect to $\phi_i,\dot{\phi}_i,\theta_i,\dot{\theta}_i$, and the B's with respect to $\phi_i,\phi_j,\theta_i,\theta_j$. In so doing, we make use of the fact that *products* of sines and cosines can always be rewritten as *sums* of sines and cosines of suitable arguments. What is meant by *linearizing* $f(\theta)$ with respect to θ? By definition,

$$\mathrm{lin}[f(\theta)]=\tfrac{1}{2}\langle f(\theta)+f(-\theta)\rangle + \tfrac{1}{2}\theta\langle (d/d\theta)[f(\theta)-f(-\theta)]\rangle \ . \tag{44}$$

In other words, we replace the even part of $f(\theta)$ by a suitable average; we replace the odd part by θ times the average of the derivative of the odd part. In the present calculation, the averaging is with respect to time and with respect to the random phases Φ_k appearing in ϕ_i and θ_i. This linearization approximation eliminates the possibility of calculating lifetime effects associated with the magnon excitation energies. Nevertheless, the approximation is essential for being able to carry the present calculation to completion.

For the angles we are considering, we can write

$$\langle\theta^{2n}\rangle=[(2n)!/n!2^n]\langle\theta^2\rangle^n \ , \tag{45}$$

$$\langle\cos\theta\rangle=\sum_{n=0}^{\infty}[(-1)^n/(2n)!]\langle\theta^{2n}\rangle=\exp(-\tfrac{1}{2}\langle\theta^2\rangle) \ , \tag{46}$$

$$\langle\phi_i^2\rangle=\langle\theta_i^2\rangle=\tfrac{1}{2}\sum_k C_k^2=N^{-1}\sum_k 2f_k \ , \tag{47}$$

$$\langle\dot{\theta}_i\phi_i\rangle=-\langle\dot{\phi}_i\theta_i\rangle=\tfrac{1}{2}\sum_k\omega_k C_k^2=N^{-1}\sum_k 2\omega_k f_k \ , \tag{48}$$

$$\begin{aligned}\mathrm{lin}(\dot{\theta}_i\sin\phi_i)&=\langle\dot{\theta}_i\sin\phi_i\rangle\\ &=\langle\dot{\theta}_i\phi_i\rangle\langle(d/d\phi_i)\sin\phi_i\rangle\\ &=\left[N^{-1}\sum_k 2\omega_k f_k\right]\exp\left[-N^{-1}\sum_k f_k\right] \ ,\end{aligned} \tag{49}$$

$$\begin{aligned}\mathrm{lin}(\dot{\theta}_i\cos\phi_i)&=\dot{\theta}_i\langle\cos\phi_i\rangle+\phi_i\langle\dot{\theta}_i(\partial/\partial\phi_i)\cos\phi_i\rangle\\ &=\left[\dot{\theta}_i-\left[N^{-1}\sum_k 2\omega_k f_k\right]\phi_i\right]\\ &\quad\times\exp\left[-N^{-1}\sum_k f_k\right] \ .\end{aligned} \tag{50}$$

After rewriting products of sines and cosines as sums of sines and cosines, the angular arguments of the resultant trigonometric functions all have the form

$$\theta=(\nu_1\phi_j+\nu_2\phi_i+\nu_3\theta_j+\nu_4\theta_i),\quad \nu_n=0,\pm 1\ . \tag{51}$$

Thus

$$\theta=\sum_k C_k(\{\nu_1\sin[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]+\nu_3\cos[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]+\nu_4\}\cos(\vec{k}\cdot\vec{R}_i-\omega_k t+\Phi_k)$$
$$+\{\nu_1\cos[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]-\nu_3\sin[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]+\nu_2\}\sin(\vec{k}\cdot\vec{R}_i-\omega_k t+\Phi_k))\ , \tag{52}$$

$$\tfrac{1}{2}\langle\theta^2\rangle=\tfrac{1}{4}\sum_k C_k^2(\{\nu_1\sin[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]+\nu_3\cos[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]+\nu_4\}^2+\{\nu_1\cos[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]-\nu_3\sin[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]+\nu_2^2\}^2)$$
$$=N^{-1}\sum_k f_k\{(\nu_1^2+\nu_2^2+\nu_3^2+\nu_4^2)+2(\nu_1\nu_2+\nu_3\nu_4)\cos[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]\}\ . \tag{53}$$

We have

$$\text{lin}(\cos\theta)=\langle\cos\theta\rangle=\exp(-\tfrac{1}{2}\langle\theta^2\rangle)\ ,$$
$$\text{lin}(\sin\theta)=\theta\langle\cos\theta\rangle=\theta\exp(-\tfrac{1}{2}\langle\theta^2\rangle)\ . \tag{54}$$

Defining the quantities

$$b\equiv N^{-1}\sum_k 2f_k=2n_0\ ,$$
$$d\equiv N^{-1}\sum_k 2\omega_k f_k\ , \tag{55}$$
$$g_{ij}\equiv N^{-1}\sum_k 2f_k\{1-\cos[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]\}\ ,$$

we can now write the linearized versions of the A's and the B's as

$$A_{ix}=de^{-(1/2)b}\ , \tag{56}$$

$$A_{iy}=\tfrac{1}{2}\sum_k C_k\sin(\vec{k}\cdot\vec{R}_i-\omega_k t+\Phi_k)$$
$$\times[(1+\epsilon)\omega_k+(1-\epsilon)(\omega_k-d)e^{-(1/2)b}]\ , \tag{57}$$

$$A_{iz}=-\tfrac{1}{2}\sum_k C_k\cos(\vec{k}\cdot\vec{R}_i-\omega_k t+\Phi_k)$$
$$\times[(1-\epsilon)\omega_k+(1+\epsilon)(\omega_k-d)e^{-(1/2)b}]\ , \tag{58}$$

$$B_{ijxx}=\tfrac{1}{2}[e^{-g_{ij}}(1+e^{-g_{ij}})-e^{-2b}(e^{+g_{ij}}-1)]\ , \tag{59}$$

$$B_{ijxy}=-\tfrac{1}{2}(1+\epsilon)(\phi_j-\phi_i)e^{-g_{ij}-(1/2)b}$$
$$-\tfrac{1}{4}(1-\epsilon)[(\phi_j-\phi_i)(1+e^{-g_{ij}})e^{-g_{ij}}$$
$$-(\phi_j+\phi_i)(e^{+g_{ij}}-1)e^{-2b}]\ , \tag{60}$$

$$B_{ijxz}=\tfrac{1}{4}(1+\epsilon)[(\theta_j-\theta_i)(1+e^{-g_{ij}})e^{-g_{ij}}$$
$$-(\theta_j+\theta_i)(e^{+g_{ij}}-1)e^{-2b}]$$
$$+\tfrac{1}{2}(1-\epsilon)(\theta_j-\theta_i)e^{-g_{ij}-(1/2)b}\ , \tag{61}$$

$$B_{ijyx}=\tfrac{1}{2}(1+\epsilon)(\phi_j-\phi_i)e^{-g_{ij}-(1/2)b}$$
$$+\tfrac{1}{4}(1-\epsilon)[(\phi_j-\phi_i)(1+e^{-g_{ij}})e^{-g_{ij}}$$
$$+(\phi_j+\phi_i)(e^{+g_{ij}}-1)e^{-2b}]\ , \tag{62}$$

$$B_{ijyy}=\tfrac{1}{2}(1+\epsilon)e^{-g_{ij}}$$
$$+\tfrac{1}{4}(1-\epsilon)[(1+e^{-g_{ij}})e^{-g_{ij}}+(e^{+g_{ij}}-1)e^{-2b}]\ , \tag{63}$$

$$B_{ijyz}=B_{ijzy}=0\ , \tag{64}$$

$$B_{ijzx}=-\tfrac{1}{4}(1+\epsilon)[(\theta_j-\theta_i)(1+e^{-g_{ij}})e^{-g_{ij}}$$
$$+(\theta_j+\theta_i)(e^{+g_{ij}}-1)e^{-2b}]$$
$$-\tfrac{1}{2}(1-\epsilon)(\theta_j-\theta_i)e^{-g_{ij}-(1/2)b}\ , \tag{65}$$

$$B_{ijzz}=\tfrac{1}{4}(1+\epsilon)[(1+e^{-g_{ij}})e^{-g_{ij}}+(e^{+g_{ij}}-1)e^{-2b}]$$
$$+\tfrac{1}{2}(1-\epsilon)e^{-g_{ij}}\ . \tag{66}$$

Considering now the final rotational tranformation [Eq. (18)], we have

$$H_{ix3}=-\Omega+H_{ix2}\ ,$$
$$H_{iy3}=H_{iy2}\cos(\Omega t)-H_{iz2}\sin(\Omega t)\ , \tag{67}$$
$$H_{iz3}=H_{iy2}\sin(\Omega t)+H_{iz2}\cos(\Omega t)\ .$$

As already announced, we wish to find a "static" solution in the $x_3y_3z_3$ coordinate system. In the absence of magnons, we took the ground state such that all spins are pointing along the x axis. In the presence of magnons, all spins are pointing along the x_3 axis. Thus, for all i,

$$S_{ix3}=\tfrac{1}{2},\quad S_{iy3}=S_{iz3}=0\ . \tag{68}$$

In order that the equations of motion be satisfied, we must have

$$H_{iy3}=H_{iz3}=0\ . \tag{69}$$

These equations are equivalent to

$$S_{ix2}=\tfrac{1}{2},\quad S_{iy2}=S_{iz2}=0\ , \tag{70}$$

$$H_{iy2}=H_{iz2}=0\ . \tag{71}$$

Equation (71) implies

$$2\hbar A_{iy}+\sum_j J_{ij}B_{ijyx}=2\hbar A_{iz}+\sum_j J_{ij}B_{ijzx}=0 . \quad (72)$$

For arbitrary ϵ, Eqs. (72) have no solution. However, for the case $\epsilon=0$ (symmetrized product of the two oscillatory transformations), we have

$$2\hbar A_{iy}+\sum_j J_{ij}B_{ijyx}=\sum_k C_k D_{ki}\sin(\vec{k}\cdot\vec{R}_i-\omega_k t+\Phi_k) ,$$
$$2\hbar A_{iz}+\sum_j J_{ij}B_{ijzx}=-\sum_k C_k D_{ki}\cos(\vec{k}\cdot\vec{R}_i-\omega_k t+\Phi_k) , \quad (73)$$

where

$$D_{ki}\equiv\hbar\omega_k(1+e^{-(1/2)b})-\hbar d e^{-(1/2)b}-\tfrac{1}{4}\sum_j J_{ij}\{1-\cos[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]\}e^{-g_{ij}}(1+e^{-g_{ij}}+2e^{-(1/2)b})$$
$$+\tfrac{1}{4}\sum_j J_{ij}\{1+\cos[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]\}e^{-2b}(e^{+g_{ij}}-1) . \quad (74)$$

It follows that Eqs. (72) are solved by setting

$$\epsilon=0,\ \ D_{ki}=0 . \quad (75)$$

D_{ki} can be made to vanish by setting

$$\hbar\omega_k=\tfrac{1}{2}\sum_j J_{ij}(\alpha_{0ij}+\alpha_{1ij}\{1-\cos[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]\}) , \quad (76)$$

where

$$\alpha_{1ij}\equiv\tfrac{1}{2}(1+e^{-(1/2)b})^{-1}[e^{-g_{ij}}(1+e^{-g_{ij}}+2e^{-(1/2)b})+e^{-2b}(e^{+g_{ij}}-1)] , \quad (77)$$

and α_{0ij} is chosen such that

$$\tfrac{1}{2}\sum_j J_{ij}\alpha_{0ij}=(e^{+(1/2)b}+1)^{-1}\hbar d-\tfrac{1}{2}(1+e^{-(1/2)b})^{-1}e^{-2b}\sum_j J_{ij}(e^{+g_{ij}}-1) . \quad (78)$$

From the definition of d, we have

$$\hbar d=\tfrac{1}{2}\sum_j J_{ij}(\alpha_{0ij}b+\alpha_{1ij}g_{ij}) . \quad (79)$$

Thus we can choose

$$\alpha_{0ij}\equiv(1-b+e^{+(1/2)b})^{-1}\times[\alpha_{1ij}g_{ij}-e^{-(3/2)b}(e^{+g_{ij}}-1)] . \quad (80)$$

Let us now go beyond the "static" state we have been considering. Rather than the values of Eq. (68), we take

$$S_{ix3}=S_{ix30}=\tfrac{1}{2},\ \ S_{iy3}=\delta S_{iy3},\ \ S_{iz3}=\delta S_{iz3} . \quad (81)$$

Correspondingly, we have

$$H_{ix3}=H_{ix30}+\delta H_{ix3},\ \ H_{iy3}=\delta H_{iy3},\ \ H_{iz3}=\delta H_{iz3} . \quad (82)$$

Making use of

$$B_{ijyy}=B_{ijzz},\ \ B_{ijyz}=B_{ijzy}=0 , \quad (83)$$

we have

$$\delta H_{iy3}=\hbar^{-1}\sum_j J_{ij}B_{ijyy}\delta S_{iy3} ,$$
$$\delta H_{iz3}=\hbar^{-1}\sum_j J_{ij}B_{ijyy}\delta S_{jz3} . \quad (84)$$

Also,

$$H_{ix30}=A_{ix}-\Omega+(2\hbar)^{-1}\sum_j J_{ij}B_{ijxx} . \quad (85)$$

Our exact equations of motion are

$$\frac{dS_{ix3}}{dt}=S_{iy3}H_{iz3}-S_{iz3}H_{iy3} ,$$
$$\frac{dS_{iy3}}{dt}=S_{iz3}H_{ix3}-S_{ix3}H_{iz3} , \quad (86)$$
$$\frac{dS_{iz3}}{dt}=S_{ix3}H_{iy3}-S_{iy3}H_{ix3} .$$

We choose to *linearize* these equations of motion with respect to the $\delta\vec{S}_i$ and $\delta\vec{H}_i$. Thus we obtain

$$\frac{dS_{ix3}}{dt}=0 ,$$
$$\frac{dS_{iy3}}{dt}=H_{ix30}\delta S_{iz3}-S_{ix30}\delta H_{iz3} , \quad (87)$$
$$\frac{dS_{iz3}}{dt}=-H_{ix30}\delta S_{iy3}+S_{ix30}\delta H_{iy3} .$$

Note that the linearization has caused δH_{ix3} to drop out of the equations of motion. We define

$$\delta S_i\equiv\delta S_{iy3}+i\delta S_{iz3} , \quad (88)$$

$$\delta H_i\equiv\delta H_{iy3}+i\delta H_{iz3}=\hbar^{-1}\sum_j J_{ij}B_{ijyy}\delta S_j , \quad (89)$$

$$E_0\equiv\hbar A_{ix}-\hbar\Omega+\tfrac{1}{2}\sum_j J_{ij}(B_{ijxx}-B_{ijyy}) , \quad (90)$$

$$\alpha_{2ij}\equiv B_{ijyy} . \quad (91)$$

Thus we can write

$$i\hbar\frac{d}{dt}\delta S_i=E_0\delta S_i+\tfrac{1}{2}\sum_j J_{ij}\alpha_{2ij}(\delta S_i-\delta S_j) . \quad (92)$$

By setting, for all i,

$$\delta S_i=C_k e^{i(\vec{k}\cdot\vec{R}_i-\omega_k t+\Phi_k)} , \quad (93)$$

we obtain

$$\hbar\omega_k=E_0+\tfrac{1}{2}\sum_j J_{ij}\alpha_{2ij}\{1-\cos[\vec{k}\cdot(\vec{R}_j-\vec{R}_i)]\} . \quad (94)$$

At this point, we choose Ω such that E_0 equals the $\hbar\omega_0$ of Eq. (76). Thus

$$\Omega \equiv A_{ix} + (2\hbar)^{-1} \sum_j J_{ij}(B_{ijxx} - B_{ijyy} - \alpha_{0ij}) = (2\hbar)^{-1} \sum_j J_{ij}[\tfrac{1}{4}(e^{-2b} - e^{-2g_{ij}})(e^{+g_{ij}} - 1) - (1 + e^{+(1/2)b})^{-1}\alpha_{0ij}] , \tag{95}$$

so that

$$\hbar\omega_k = \tfrac{1}{2} \sum_j J_{ij}\{\alpha_{0ij} + \alpha_{2ij}[1 - \cos\vec{k}\cdot(\vec{R}_j - \vec{R}_i)]\} . \tag{96}$$

In order to understand the results we have obtained, we expand α_{0ij}, α_{1ij}, α_{2ij}, and Ω in powers of b and g_{ij}. Terms proportional to $b^{n_1} g_{ij}^{n_2}$ result from n-magnon interactions, where $n = (n_1 + n_2 + 1)$:

$$\alpha_{0ij} = \tfrac{3}{4} g_{ij}(b - g_{ij}) - \tfrac{1}{48} g_{ij}(18b^2 + 3bg_{ij} - 20g_{ij}^2) + \cdots , \tag{97}$$

$$\alpha_{1ij} = 1 - g_{ij} + \tfrac{1}{2} g_{ij}(2g_{ij} - b) + \tfrac{1}{8} g_{ij}(3b^2 + bg_{ij} - \tfrac{10}{3} g_{ij}^2) + \cdots , \tag{98}$$

$$\alpha_{2ij} = 1 - g_{ij} + \tfrac{1}{2} g_{ij}(2g_{ij} - b) + \tfrac{1}{12} g_{ij}(6b^2 - 3bg_{ij} - 5g_{ij}^2) + \cdots , \tag{99}$$

$$\hbar\Omega = -\tfrac{1}{4} \sum_j J_{ij}[g_{ij}(b - g_{ij}) + g_{ij}(5b^2 - 7bg_{ij} + 2g_{ij}^2) + \cdots] . \tag{100}$$

To the accuracy of two-magnon interactions, $\alpha_{1ij} = \alpha_{2ij} = (1 - g_{ij})$ and $\alpha_{0ij} = \Omega = 0$. This is just the form of $\hbar\omega_k$ derived by Bloch[7] and by Poling and Parmenter,[8] known to lead to the correct low-temperature properties. Most remarkably, $\alpha_{1ij} = \alpha_{2ij}$ to the accuracy of three-magnon interactions. The two alternative ways of calculating $\hbar\omega_k$ lead to different results only in the presence of four-magnon interactions. A gap first appears in the magnon excitation spectrum in the presence of three-magnon interactions. To this accuracy, the gap is

$$\hbar\omega_0 = \tfrac{3}{8} \sum_j J_{ij} g_{ij}(b - g_{ij}) . \tag{101}$$

This should be compared with the Δ_{01} of Eq. (8), which can be written as

$$\Delta_{01} = \tfrac{1}{4} c_1 \sum_j J_{ij} g_{ij}(1 - g_{ij}) b . \tag{102}$$

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