# Magnon Density Fluctuations in the Heisenberg Ferromagnet

GEORGE F. REITER*
*Department of Physics, University of California, Irvine, California*

(Received 12 February 1968)

It is shown, by means of a decoupling procedure for the equations of motion of a spin system governed by the Heisenberg Hamiltonian, that the distribution of magnons in phase space satisfies the quantum-mechanical Boltzmann equation at low temperatures. We conclude from this that disturbances in the longitudinal component of the magnetization, which may be thought of as density fluctuations in the gas of magnons, propagate essentially undamped at long wavelengths, in complete analogy with the propagation of sound in a gas of real particles. The interaction of this hydrodynamic mode with phonons is considered, and the damping coefficient of the phonons when their velocity coincides with the magnon sound velocity is obtained.

## I. INTRODUCTION

THE dynamical behavior of the Heisenberg ferromagnet at low temperatures has been studied extensively.[1–4] Most of this work has centered on the behavior of the spin waves, or fluctuations of the magnetization transverse to the direction of the spontaneous magnetization. The resultant picture is that the lowest excited states can be described in terms of a weakly interacting gas of particles, the magnons. It has been suggested by Gulayev,[5] on the basis of this analogy, that the density fluctuations in such a gas would behave in the same fashion as do those in a gas of real particles. That is, they would propagate as a sound wave if their wavelength were sufficiently long. Since an increase in the density of magnons in a region of the lattice will result in a decrease in the longitudinal component of the magnetization, this sound wave will manifest itself as a fluctuation in the longitudinal component of the magnetization, and will therefore appear as a pole in the longitudinal susceptibility.

There have been several attempts to calculate the longitudinal susceptibility directly from the equations of motion.[6–9] These works do not support the conclusion that there is a hydrodynamic soundlike mode, but are either incorrect or not valid in the region of wave vectors and frequencies of interest.

The present work calculates the susceptibility in the collision-dominated or hydrodynamic regime. This is accomplished by demonstrating that the distribution of magnons in phase space satisfies a Boltzmann equation when the wave vector of the fluctuation is small. One can then infer the behavior in the hydrodynamic region from the well-known properties of the solution of this equation, and conclude that the magnon sound wave does in fact exist.

## II. LONGITUDINAL SUSCEPTIBILITY—COLLISIONLESS REGIME

First, we shall discuss the behavior in the limit that collisions are neglected. From the Heisenberg Hamiltonian for a lattice of spins,

$$\mathcal{H}=-\hbar H\sum_i S_i^z-\tfrac{1}{2}\hbar\sum_{i,j}V_{ij}\mathbf{S}_i\cdot\mathbf{S}_j,$$

the commutation relations for the spin operators

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

and the Heisenberg equation of motion for an operator $A$,

$$i\hbar(\partial A/\partial t)=[A,\mathcal{H}],$$

we can obtain a chain of equations describing the temporal behavior of the magnetization and the various correlation functions. In terms of the Fourier-transformed variables

$$\mathbf{S}(\mathbf{q})=N^{-1/2}\sum_i\exp(-i\mathbf{q}\cdot\mathbf{r}_i)\mathbf{S}_i,$$
$$V(\mathbf{q})=\sum_i\exp[i\mathbf{q}\cdot(\mathbf{r}_i-\mathbf{r}_j)]V_{ij},$$

the equations of motion can be written

$$i(\partial/\partial t)\langle S^z(\mathbf{q})\rangle=\tfrac{1}{2}N^{-1/2}\sum_{\mathbf{q}'}[V(\mathbf{q}-\mathbf{q}')-V(\mathbf{q}')]\times\langle S^-(\mathbf{q}')S^+(\mathbf{q}-\mathbf{q}')\rangle,\quad(1a)$$

$$\begin{aligned}i(\partial/\partial t)\langle S^-(\mathbf{q}_1)S^+(\mathbf{q}_2)\rangle&=[\omega(\mathbf{q}_2)-\omega(\mathbf{q}_1)]\langle S^-(\mathbf{q}_1)S^+(\mathbf{q}_2)\rangle\\&+N^{-1/2}\sum_{\mathbf{q}'}V(\mathbf{q}')-V(\mathbf{q}_1-\mathbf{q}')\\&\times\langle S^-(\mathbf{q}_1-\mathbf{q}')[S^z(\mathbf{q}')-N^{1/2}S\delta(\mathbf{q}')]S^+(\mathbf{q}_2)\rangle\\&-N^{1/2}\sum_{\mathbf{q}'}V(\mathbf{q}')-V(\mathbf{q}_2-\mathbf{q}')\\&\times\langle S^-(\mathbf{q}_1)[S^z(\mathbf{q}')-N^{1/2}S\delta(\mathbf{q}')]S^+(\mathbf{q}_2-\mathbf{q}')\rangle,\quad(1b)\end{aligned}$$

where $\omega(\mathbf{q})=H+S[V(0)-V(\mathbf{q})]$.

* This work is based in part upon a thesis submitted to Stanford University.
[1] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).
[2] F. Dyson, Phys. Rev. **102**, 1217 (1956).
[3] M. Wortis, Phys. Rev. **132**, 1522 (1965).
[4] J. F. Cooke and H. A. Gersch, Phys. Rev. **153**, 641 (1967).
[5] Yu. V. Gulayev, Zh. Eksperim. i Teor. Fiz., Pis'ma v Redaktsiyu **2**, 3 (1965) [English transl.: Soviet Phys.—JETP Letters **2**, 1 (1965)].
[6] H. Mori and K. Kawasaki, Progr. Theoret. Phys. (Kyoto) **27**, 529 (1962).
[7] H. Mori, *Many Body Theory* (W. A. Benjamin, Inc., New York, 1966).
[8] S. H. Liu, Phys. Rev. **139**, 1522 (1965).
[9] A. I. Mitsek, Fiz. Tverd. Tela. **8**, 1498 (1966) [English transl.: Soviet Phys.—Solid State **8**, 1191 (1966)].

The averages $\langle\ \rangle$ are to be taken with respect to some initial density matrix. Since we are interested in the linear response, we will take this to be of the form $\rho_{eq}+\Delta\rho$, where $\rho_{eq}$ is the thermal-equilibrium density matrix, and $\Delta\rho$ is the initial perturbation, assumed to be small. Averages over $\rho_{eq}$ will be denoted by $\langle\langle\ \rangle\rangle$. We have suppressed the time dependence of the averages for simplicity of notation.

Equations (1a) and (1b) can be given a suggestive semiclassical interpretation. A classical gas can be described in terms of the density of particles with momentum $\mathbf{P}$ at the point $\mathbf{X}$. The quantum-mechanical analog of this distribution is the Wigner function,[10] which can be defined as

$$f(\mathbf{X},\mathbf{P})=(2\pi)^{-3}\int \exp(i\mathbf{n}\cdot\mathbf{X}) \times[\mathrm{Tr}\rho a^\dagger(\mathbf{P}+\tfrac{1}{2}(\hbar\mathbf{n}))a(\mathbf{P}-\tfrac{1}{2}(\hbar\mathbf{n}))]d^3\eta,$$

where $a^\dagger(\mathbf{P})$ is the creation operator for a particle of momentum $\mathbf{P}$. Thus the spatial Fourier transform of this quantity can be written as

$$\mathrm{Tr}\rho a^\dagger(\mathbf{P}+\hbar\,\tfrac{1}{2}\mathbf{q})a(\mathbf{P}-\hbar\,\tfrac{1}{2}\mathbf{q}).$$

At low temperatures, $S^-(\mathbf{q})$ behaves like a creation operator for a magnon of momentum $\hbar\mathbf{q}$, $S^+(-\mathbf{q})$ as a destruction operator. Hence, if we choose $\mathbf{q}_1=\mathbf{q}'+\frac{1}{2}\mathbf{q}$ $\mathbf{q}_2=\frac{1}{2}\mathbf{q}-\mathbf{q}'$, we can interpret Eq. (1b) as the equation of motion for the Wigner function.

If the Wigner function is summed over the momentum variable, the resultant function gives the density of particles at $\mathbf{X}$. A similar relation holds for its spin analog if $S=\frac{1}{2}$. We have in this case

$$S_i^z=S-(S_i^-S_i^+/2S)$$

or, in terms of the Fourier-transformed variables,

$$S^z(\mathbf{q})-N^{1/2}S\delta(\mathbf{q}) =-(N^{-1/2}/2S)\sum_{\mathbf{q}'}S^-(\mathbf{q}'+\tfrac{1}{2}\mathbf{q})S^+(\tfrac{1}{2}\mathbf{q}-\mathbf{q}').\quad(2)$$

Thus, if we interpret $N^{1/2}S\delta(\mathbf{q})-\langle S^z(\mathbf{q})\rangle$ as the Fourier transform of the density of particles, we will be consistent with the interpretation of

$$(1/2S)\langle S^-(\mathbf{q}'+\tfrac{1}{2}\mathbf{q})S^+(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')\rangle$$

as the Fourier transform of the distribution of magnons in phase space. Equation (1a) can then be interpreted as the continuity equation. We should emphasize that these remarks are only intended to facilitate the interpretation of the equations. We will not make use of any transformation to Bose operators, so that the quantities we will deal with will not be strictly Wigner functions. Also, the restriction that $S=\frac{1}{2}$ is not necessary and will be removed at the end of the calculation.

Using (2) we can write (1b) as

$$\begin{aligned}i(\partial/\partial t)&\langle S^-(\mathbf{q}_1)S^+(-\mathbf{q}_2)\rangle\\ &=[\omega(\mathbf{q}_2)-\omega(\mathbf{q}_1)]\langle S^-(\mathbf{q}_1)S^+(-\mathbf{q}_2)\rangle\\ &+(2SN)^{-1}\sum_{\mathbf{q}_1'\mathbf{q}_2'\mathbf{q}_3'}\delta(\mathbf{q}_1+\mathbf{q}_3'-\mathbf{q}_1'-\mathbf{q}_2')\Gamma(\mathbf{q}_1\mathbf{q}_3';\mathbf{q}_1'\mathbf{q}_2')\\ &\quad\times\langle S^-(\mathbf{q}_1')S^-(\mathbf{q}_2')S^+(-\mathbf{q}_3')S^+(-\mathbf{q}_2)\rangle\\ &-(2SN)^{-1}\sum_{\mathbf{q}_2'\mathbf{q}_3'\mathbf{q}_4'}\delta(\mathbf{q}_2+\mathbf{q}_2'-\mathbf{q}_3'-\mathbf{q}_4')\Gamma(\mathbf{q}_2\mathbf{q}_2';\mathbf{q}_3'\mathbf{q}_4')\\ &\quad\times\langle S^-(\mathbf{q}_1)S^-(\mathbf{q}_2')S^+(-\mathbf{q}_3')S^+(-\mathbf{q}_4')\rangle,\quad(3)\end{aligned}$$

where

$$\Gamma(\mathbf{q}_1\mathbf{q}_2;\mathbf{q}_3\mathbf{q}_4) =\tfrac{1}{2}[V(\mathbf{q}_1-\mathbf{q}_3)+V(\mathbf{q}_1-\mathbf{q}_4)-V(\mathbf{q}_3)-V(\mathbf{q}_4)].$$

The quantity $\Gamma$ is the matrix element that appears in the four-magnon interaction term when the Heisenberg Hamiltonian is converted to a boson Hamiltonian by means of the Dyson-Maleev transformation.[11]

At this point we can make an approximation, similar to the Hartree-Fock approximation for the electron gas, and convert the identity (3) into an equation from which we may determine $\langle S^-(\mathbf{q}_1)S^+(\mathbf{q}_2)\rangle$:

$$\begin{aligned}\langle S^-(\mathbf{q}_1)&S^-(\mathbf{q}_2)S^+(\mathbf{q}_3)S^+(\mathbf{q}_4)\rangle\\ &\simeq\langle S^-(\mathbf{q}_1)S^+(\mathbf{q}_3)\rangle\langle S^-(\mathbf{q}_2)S^+(\mathbf{q}_4)\rangle\\ &\quad+\langle S^-(\mathbf{q}_1)S^+(\mathbf{q}_4)\rangle\langle S^-(\mathbf{q}_2)S^+(\mathbf{q}_3)\rangle.\quad(4)\end{aligned}$$

The justification for this approximation is the observation that any physically realizable density matrix must have the cluster property. That is, any two groups of spins must behave independently as the distance between the two groups is increased. Thus the quantity $\langle S_i^-S_j^-S_k^+S_l^+\rangle$ must approach $\langle S_i^-S_k^+\rangle\langle S_j^-S_l^+\rangle$ as the pair $[i,k]$ recedes from the pair $[j,l]$. Quite generally, the average of a product of operators can be written as a sum over all possible decompositions into subsets, plus a term for which no decomposition is made. This latter term will have the property of vanishing unless all the operators that enter the product refer to sites that are close to one another. Our approximation consists of neglecting this term entirely. We need not consider factors such as $\langle S_i^-S_j^-\rangle\langle S_k^+S_l^+\rangle$ since the initial perturbation will be chosen so that these terms vanish initially. The rotational invariance of the Hamiltonian ensures that they will vanish for all time.

The equation that results from substituting (4) into (3) is nonlinear, and we do not propose to solve it. We note that

$$(1/2S)\langle S^-(\mathbf{q}_1)S^+(\mathbf{q}_2)\rangle=\delta(\mathbf{q}_1+\mathbf{q}_2)\bar{n}(\mathbf{q}_1)$$

is a stationary solution of this equation for any value of $\bar{n}$. This follows from the translational invariance.

[10] H. Mori, I. Oppenheim, and J. Ross, *Studies in Statistical Mechanics* (North-Holland Publishing Co., Amsterdam, 1965), Vol. I.

[11] S. V. Maleev, Zh. Eksperim. i Teor. Fiz. **33**, 1010 (1957) [English transl.: Soviet Phys.—JETP **6**, 776 (1958)].

We choose $\bar{n}$ to be the equilibrium correlation function, $\langle\langle S^-(\mathbf{q})S^+(-\mathbf{q})\rangle\rangle$, and linearize the equations about this stationary solution. That is, we make the replacement

$$(1/2S)\langle S^-(\mathbf{q}'+\tfrac{1}{2}\mathbf{q})S^+(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')\rangle =\delta(\mathbf{q})\bar{n}(\mathbf{q}')+\epsilon f(\mathbf{q},\mathbf{q}')$$

and drop all terms of order $\epsilon^2$. The resulting equation, (5), is linear and describes the behavior of small deviations of the magnon distribution function from its equilibrium value:

$$\begin{aligned} i(\partial/\partial t)f(\mathbf{q},\mathbf{q}') &= [\omega_1(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')-\omega_1(\tfrac{1}{2}\mathbf{q}+\mathbf{q}')]f(\mathbf{q},\mathbf{q}') \\ &+N^{-1}\sum_{\mathbf{q}''}[(V(\mathbf{q}'+\tfrac{1}{2}\mathbf{q}-\mathbf{q}'')-V(\mathbf{q}''))\bar{n}(\tfrac{1}{2}\mathbf{q}+\mathbf{q}') \\ &\quad -(V(\mathbf{q}'+\tfrac{1}{2}\mathbf{q}-\mathbf{q}'')-V(\mathbf{q}-\mathbf{q}'')) \\ &\quad \times\bar{n}(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')]\langle S^-(\mathbf{q}'')S^+(\mathbf{q}-\mathbf{q}'')\rangle \\ &+N^{-1/2}[(V(\mathbf{q})-V(\tfrac{1}{2}\mathbf{q}+\mathbf{q}'))\bar{n}(\tfrac{1}{2}\mathbf{q}+\mathbf{q}') \\ &-(V(\mathbf{q})-V(\tfrac{1}{2}\mathbf{q}-\mathbf{q}'))\bar{n}(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')]\delta\langle S^z(\mathbf{q})\rangle. \quad (5)\end{aligned}$$

$\omega_1(\mathbf{q})$ is a renormalized magnon energy given by

$$\omega_1(\mathbf{q})=H+S[V(0)-V(\mathbf{q})] -2N^{-1}\sum_{\mathbf{q}'}\Gamma(\mathbf{q},\mathbf{q}';\mathbf{q},\mathbf{q}')\bar{n}(\mathbf{q}')$$

and $\delta\langle S^z(\mathbf{q})\rangle$ is $\langle S^z(\mathbf{q})\rangle-\langle\langle S^z(\mathbf{q})\rangle\rangle$. If we ignore the sum on the right of Eq. (5), the solution can readily be obtained. Introducing the Laplace-transformed quantities

$$\tilde{f}(\mathbf{q},\mathbf{q}')=\int_0^\infty e^{izt}f(\mathbf{q},\mathbf{q}',t)dt,$$

$$\tilde{\rho}(\mathbf{q})=\int_0^\infty e^{izt}\delta\langle S^z(\mathbf{q},t)\rangle dt,$$

we have

$$\tilde{f}(\mathbf{q},\mathbf{q}')=\frac{if(\mathbf{q},\mathbf{q}',t=0)}{z-\omega_1(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')+\omega_1(\tfrac{1}{2}\mathbf{q}+\mathbf{q}')} +N^{-1/2}\frac{[(V(\mathbf{q})-V(\tfrac{1}{2}\mathbf{q}+\mathbf{q}'))\bar{n}(\tfrac{1}{2}\mathbf{q}+\mathbf{q}')-(V(\mathbf{q})-V(\tfrac{1}{2}\mathbf{q}-\mathbf{q}'))\bar{n}(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')]}{z-\omega_1(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')+\omega_1(\tfrac{1}{2}\mathbf{q}+\mathbf{q}')}\tilde{\rho}(\mathbf{q}). \quad (6)$$

We have left the initial condition unspecified until now. An appropriate choice for the perturbation $\Delta\rho$ is

$$\Delta\rho=-i\epsilon[S^z(-\mathbf{q}),\rho_{\text{eq}}]. \quad (7)$$

$\tilde{\rho}(\mathbf{q})$ is then precisely the longitudinal susceptibility, $\chi(\mathbf{q},z)$, defined by

$$\chi(\mathbf{q},z)=-i\int_0^\infty e^{izt}\langle\langle[S^z(\mathbf{q},t),S^z(-\mathbf{q})]\rangle\rangle dt \quad \text{Im}z>0.$$

Using (7) and (2), we obtain an expression for $\chi(\mathbf{q},z)$:

$$\chi(\mathbf{q},z)=-N^{-1}\sum_{q'}\frac{\bar{n}(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')-\bar{n}(\tfrac{1}{2}\mathbf{q}+\mathbf{q}')}{z-\omega_1(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')+\omega_1(\tfrac{1}{2}\mathbf{q}+\mathbf{q}')} \times\left(1+N^{-1}\sum_{q'}\frac{\Delta(\mathbf{q},\mathbf{q}')}{z-\omega_1(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')+\omega_1(\tfrac{1}{2}\mathbf{q}+\mathbf{q}')}\right)^{-1}, \quad (8)$$

where

$$\Delta(\mathbf{q},\mathbf{q}')=V(\mathbf{q}'-\tfrac{1}{2}\mathbf{q})\bar{n}(\mathbf{q}'-\tfrac{1}{2}\mathbf{q})-V(\mathbf{q}'+\tfrac{1}{2}\mathbf{q})\bar{n}(\mathbf{q}'+\tfrac{1}{2}\mathbf{q}) -V(\mathbf{q})(\bar{n}(\mathbf{q}'-\tfrac{1}{2}\mathbf{q})-\bar{n}(\mathbf{q}'+\tfrac{1}{2}\mathbf{q})).$$

## III. DISCUSSION OF SUSCEPTIBILITY IN THE COLLISIONLESS LIMIT

The numerator in (8) is essentially the susceptibility appropriate to a noninteracting gas of bosons. The denominator gives the corrections arising from the average field experienced by a magnon as a result of the interactions. The zeros of the analytic continuation of the denominator into the lower half of the frequency plane correspond to collective excitations of the system, if they lie sufficiently close to the real axis. It is important to note that there are no such zeros. In particular, for $\mathbf{q}$ sufficiently small that the finite differences appearing in (8) may be replaced by derivatives, we show in the Appendix that the real and imaginary parts of the sum in the denominator are less than $S-\langle\langle S^z\rangle\rangle/2S$ and $(S-\langle\langle S^z\rangle\rangle)/S$, respectively, for all values of $z$ on the real axis. Thus, when $(S-\langle\langle S^z\rangle\rangle)\ll 2S$, the sum in the denominator is uniformly small compared to unity. Therefore, there is no collective mode and it is a good approximation to neglect the effect of the average field on the motion of the magnons. This is to be expected, since the effective interaction between magnons is short ranged.

The effect of the term that we have neglected in (5) would be to add to the numerator and to the sum in the denominator terms proportional to $(\bar{n})^2$. These will be smaller by a factor of $S-\langle\langle S^z\rangle\rangle$ than the terms we have kept, and do not affect the conclusion we have reached. This conclusion agrees with the results of Liu,[8] whose expression for the susceptibility at low temperatures differs from ours essentially in the definition of

$\Delta(\mathbf{q}, \mathbf{q}')$. Liu's expression is related to ours by

$$\Delta^L(\mathbf{q}, \mathbf{q}') = \Delta(\mathbf{q}, \mathbf{q}') + (V(\mathbf{q}'+\tfrac{1}{2}\mathbf{q}) - V(\mathbf{q}'-\tfrac{1}{2}\mathbf{q}))n(\mathbf{q}'+\tfrac{1}{2}\mathbf{q}).$$

Since Liu's result is based on the random phase approximation, which neglects the dynamical correlations, we expect that our result will be more accurate, although the corrections to the noninteracting susceptibility are negligible in either case.

Our results contradict the work of Mitsek,[9] whose decoupling scheme appears to us to be invalid below the Curie temperature, since it neglects the effect of the free drift of the magnons. As we have seen, this is the dominant term in the equations of motion for the distribution function, and may not be neglected.

In regard to the work of Mori and Kawasaki,[6,7] we note that, in the present approximation, the longitudinal magnetization does not satisfy a macroscopic equation at all, since the initial value of the magnetization does not determine its future values, even for long times. This will remain true when we take the effect of the collisions between magnons into account, since it will be $f(\mathbf{q}, \mathbf{q}')$ that satisfies an equation of motion of the type suggested by Mori and Kawasaki, not $\langle S^z(\mathbf{q})\rangle$.

We can readily see that Eq. (5) does not include the effect of collisions between the magnons, since it implies that

$$(\partial/\partial t)\langle S^-(\mathbf{q}')S^+(-\mathbf{q}')\rangle = 0$$

and hence does not describe the relaxation of the magnon velocity distribution. If $\bar{\tau}$ is an average relaxation time for the magnons, then Eq. (5) is valid only when $\omega\bar{\tau} \gg 1$. To obtain an expression for $\chi(\mathbf{q}, \omega)$ valid in the opposite limit, we must include the effect of the relaxation of $\langle S^-(\mathbf{q}')S^+(-\mathbf{q}')\rangle$ in the equations of motion.

## IV. DERIVATION OF THE BOLTZMANN EQUATION

The effect of collisions between the magnons is contained in the part of $\langle S^-(\mathbf{q}_1)S^-(\mathbf{q}_2)S^+(\mathbf{q}_3)S^+(\mathbf{q}_4)\rangle$ that we have neglected. We will calculate this directly from the equations of motion, (9). Equation (9) is exact. The second set of terms results from normal ordering the last set, where by normal ordering, we mean that all $S^+$ are to the right and all $S^-$ to the left of the $S^z$. The purpose of this reordering is to separate those terms, in the linearized equation of motion that we will eventually arrive at, that are proportional to $\bar{n}$ from those that are proportional to $(\bar{n})^2$.

$$\begin{aligned}
i(\partial/\partial t)\langle S^-(\mathbf{q}_1)S^-(\mathbf{q}_2)S^+(-\mathbf{q}_3)S^+(-\mathbf{q}_4)\rangle &= [\omega(\mathbf{q}_3)+\omega(\mathbf{q}_4)-\omega(\mathbf{q}_1)-\omega(\mathbf{q}_2)]\langle S^-(\mathbf{q}_1)S^-(\mathbf{q}_2)S^+(-\mathbf{q}_3)S^+(-\mathbf{q}_4)\rangle \\
&+N^{-1}\sum_{\mathbf{q}_1'\mathbf{q}_2'}\Gamma(\mathbf{q}_1\mathbf{q}_2;\mathbf{q}_1'\mathbf{q}_2')\delta(\mathbf{q}_1+\mathbf{q}_2-\mathbf{q}_1'-\mathbf{q}_2')\langle S^-(\mathbf{q}_1')S^-(\mathbf{q}_2')S^+(-\mathbf{q}_3)S^+(-\mathbf{q}_4)\rangle \\
&-N^{-1}\sum_{\mathbf{q}_3'\mathbf{q}_4'}\Gamma(\mathbf{q}_3\mathbf{q}_4;\mathbf{q}_3'\mathbf{q}_4')\delta(\mathbf{q}_3+\mathbf{q}_4-\mathbf{q}_3'-\mathbf{q}_4')\langle S^-(\mathbf{q}_1)S^-(\mathbf{q}_2)S^+(-\mathbf{q}_3')S^+(-\mathbf{q}_4')\rangle \\
&+(2SN)^{-1}\sum_{\mathbf{q}_1'\mathbf{q}_5\mathbf{q}_6}\Gamma(\mathbf{q}_1\mathbf{q}_6;\mathbf{q}_1'\mathbf{q}_5)\delta(\mathbf{q}_1+\mathbf{q}_6-\mathbf{q}_1'-\mathbf{q}_5)\langle S^-(\mathbf{q}_1')S^-(\mathbf{q}_2)S^-(\mathbf{q}_5)S^+(-\mathbf{q}_6)S^+(-\mathbf{q}_3)S^+(-\mathbf{q}_4)\rangle \\
&+(2SN)^{-1}\sum_{\mathbf{q}_2'\mathbf{q}_5\mathbf{q}_6}\Gamma(\mathbf{q}_2\mathbf{q}_6;\mathbf{q}_2'\mathbf{q}_5)\delta(\mathbf{q}_2+\mathbf{q}_6-\mathbf{q}_2'-\mathbf{q}_5)\langle S^-(\mathbf{q}_1)S^-(\mathbf{q}_2')S^-(\mathbf{q}_5)S^+(-\mathbf{q}_6)S^+(-\mathbf{q}_3)S^+(-\mathbf{q}_4)\rangle \\
&-(2SN)^{-1}\sum_{\mathbf{q}_3'\mathbf{q}_5\mathbf{q}_6}\Gamma(\mathbf{q}_3\mathbf{q}_5;\mathbf{q}_3'\mathbf{q}_6)\delta(\mathbf{q}_3+\mathbf{q}_5-\mathbf{q}_3'-\mathbf{q}_6')\langle S^-(\mathbf{q}_1)S^-(\mathbf{q}_2)S^-(\mathbf{q}_5)S^+(-\mathbf{q}_6)S^+(-\mathbf{q}_3')S^+(-\mathbf{q}_4)\rangle \\
&-(2SN)^{-1}\sum_{\mathbf{q}_4'\mathbf{q}_5\mathbf{q}_6}\Gamma(\mathbf{q}_4\mathbf{q}_5;\mathbf{q}_4'\mathbf{q}_6)\delta(\mathbf{q}_4+\mathbf{q}_5-\mathbf{q}_4'-\mathbf{q}_6)\langle S^-(\mathbf{q}_1)S^-(\mathbf{q}_2)S^-(\mathbf{q}_5)S^+(-\mathbf{q}_6)S^+(-\mathbf{q}_3)S^+(-\mathbf{q}_4')\rangle. \quad (9)
\end{aligned}$$

The approximation that we shall make to close Eq. (9) is

$$\begin{aligned}
\langle S^-(\mathbf{q}_1)S^-(\mathbf{q}_2)S^-(\mathbf{q}_3)S^+(\mathbf{q}_4)S^+(\mathbf{q}_5)S^+(\mathbf{q}_6)\rangle &= \langle S^-(\mathbf{q}_1)S^+(\mathbf{q}_4)\rangle\langle S^-(\mathbf{q}_2)S^+(\mathbf{q}_5)\rangle\langle S^-(\mathbf{q}_3)S^+(\mathbf{q}_6)\rangle \\
&+[\text{all permutations}]. \quad (10)
\end{aligned}$$

We define the connected part of the four-spin correlation function $C(\mathbf{q}_1\mathbf{q}_2;\mathbf{q}_3\mathbf{q}_4)$ as

$$\begin{aligned}
C(\mathbf{q}_1\mathbf{q}_2;\mathbf{q}_3\mathbf{q}_4) &= \langle S^-(\mathbf{q}_1)S^-(\mathbf{q}_2)S^+(-\mathbf{q}_3)S^+(-\mathbf{q}_4)\rangle \\
&-\langle S^-(\mathbf{q}_1)S^+(-\mathbf{q}_3)\rangle\langle S^-(\mathbf{q}_2)S^+(-\mathbf{q}_4)\rangle \\
&-\langle S^-(\mathbf{q}_1)S^+(-\mathbf{q}_4)\rangle\langle S^-(\mathbf{q}_2)S^+(-\mathbf{q}_3)\rangle. \quad (11)
\end{aligned}$$

We are primarily interested in the limit that the wave vector of the external disturbance is small, and so we will consider first the case in which it vanishes entirely. We will then have

$$(2S)^{-1}\langle S^{-}(\mathbf{q}_1)S^{+}(-\mathbf{q}_2)\rangle=\delta(\mathbf{q}_1-\mathbf{q}_2)n(\mathbf{q}_4) \tag{12}$$

and we wish to obtain an equation describing the evolution of $n(\mathbf{q})$. The equation of motion for $C(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4)$ is, from Eqs. (9)–(12),

$$\begin{aligned}
i(\partial/\partial t)(2S)^{-2}C(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4) &= (\omega(\mathbf{q}_3)+\omega(\mathbf{q}_4)-\omega(\mathbf{q}_1)-\omega(\mathbf{q}_2))(2S)^{-2}C(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4)\\
&+2\delta(\mathbf{q}_1+\mathbf{q}_2-\mathbf{q}_3-\mathbf{q}_4)\Gamma(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4)n(\mathbf{q}_3)n(\mathbf{q}_4)(1+n(\mathbf{q}_1)+n(\mathbf{q}_2))\\
&-2\delta(\mathbf{q}_1+\mathbf{q}_2-\mathbf{q}_3-\mathbf{q}_4)\Gamma(\mathbf{q}_3\mathbf{q}_4;\ \mathbf{q}_1\mathbf{q}_2)n(\mathbf{q}_1)n(\mathbf{q}_2)(1+n(\mathbf{q}_3)+n(\mathbf{q}_4))\\
&+(2S)^{-2}N^{-1}\sum_{\mathbf{q}_1'\mathbf{q}_2'}\Gamma(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_1'\mathbf{q}_2')\delta(\mathbf{q}_1+\mathbf{q}_2-\mathbf{q}_1'-\mathbf{q}_2')C(\mathbf{q}_1'\mathbf{q}_2';\ \mathbf{q}_3\mathbf{q}_4)\\
&-(2S)^{-2}N^{-1}\sum_{\mathbf{q}_3'\mathbf{q}_4'}\Gamma(\mathbf{q}_3\mathbf{q}_4;\ \mathbf{q}_3'\mathbf{q}_4')\delta(\mathbf{q}_3+\mathbf{q}_4-\mathbf{q}_3'-\mathbf{q}_4')C(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3'\mathbf{q}_4')\\
&-[\delta(\mathbf{q}_1-\mathbf{q}_3)\delta(\mathbf{q}_2-\mathbf{q}_4)+\delta(\mathbf{q}_1-\mathbf{q}_4)\delta(\mathbf{q}_2-\mathbf{q}_3)]i(\partial/\partial t)n(\mathbf{q}_3)n(\mathbf{q}_4).
\end{aligned} \tag{13}$$

We shall neglect the sums in (13). As we shall see, this corresponds to a Born approximation for the scattering amplitude of the magnons with moments $\mathbf{q}_3$ and $\mathbf{q}_4$. Since the magnons involved in the scattering process are thermally excited, they will have a combined momentum much smaller than that needed for the formation of a bound state[3] (approximately $\frac{2}{3}$ of a reciprocal-lattice vector), and this will be a reasonable approximation. Furthermore, it is the structure of the collision term that is important for our considerations, not the magnitude of the scattering amplitude.

With this approximation, Eqs. (13) and (3) may readily be solved by Laplace transforms. We require, however, the initial value of $C(\mathbf{q}_1\mathbf{q}_2;\mathbf{q}_3\mathbf{q}_4)$. This quantity is essentially irrelevant to the dynamics, and produces effects that vanish in a time which is roughly that necessary for an average magnon to traverse a distance on the order of the range of the initial correlations. We will, therefore, set this quantity to zero. The solution for the Laplace transform of $C(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4)$ is then

$$\begin{aligned}
(2S)^{-2}\tilde{C}(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4z) &= 2N^{-1}\delta(\mathbf{q}_1+\mathbf{q}_2-\mathbf{q}_3-\mathbf{q}_4)\\
&\quad+(z+\omega(\mathbf{q}_1)+\omega(\mathbf{q}_2)-\omega(\mathbf{q}_3)-\omega(\mathbf{q}_4)-\omega(\mathbf{q}_4))^{-1}\\
&\quad\times\{\Gamma(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4)\tilde{n}(\mathbf{q}_3)*\tilde{n}(\mathbf{q}_4)*[1+\tilde{n}(\mathbf{q}_1)+\tilde{n}(\mathbf{q}_2)]\\
&\quad-\Gamma(\mathbf{q}_3\mathbf{q}_4;\ \mathbf{q}_1\mathbf{q}_2)\tilde{n}(\mathbf{q}_1)*\tilde{n}(\mathbf{q}_2)*[1+\tilde{n}(\mathbf{q}_3)+\tilde{n}(\mathbf{q}_4)]\}\\
&\quad-(\delta(\mathbf{q}_1-\mathbf{q}_3)\delta(\mathbf{q}_2-\mathbf{q}_4)+\delta(\mathbf{q}_1-\mathbf{q}_4)\delta(\mathbf{q}_2-\mathbf{q}_3))\\
&\quad\times(\tilde{n}(\mathbf{q}_1)*\tilde{n}(\mathbf{q}_2)-iz^{-1}n(\mathbf{q}_1,\ t=0)n(\mathbf{q}_2,\ t=0)).
\end{aligned} \tag{14}$$

The symbol $A*B$ denotes the convolution of the transforms of $A$ and $B$.

When (14) is substituted into the Laplace-transformed version of (3), we obtain a closed equation for the quantity $\tilde{n}(\mathbf{q},\ z)$. It should be noted that as a consequence of the delta functions of momenta, the last term in (14) does not contribute to this equation. The quantity $\tilde{C}(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4z)$ appears in this equation summed over three of the momentum variables and multiplied by $\Gamma$. In such a sum, it is permissible to replace $z$ by $i\epsilon$ in the factor

$$[z-\omega(\mathbf{q}_1)+\omega(\mathbf{q}_2)-\omega(\mathbf{q}_3)-\omega(\mathbf{q}_4)]^{-1}.$$

This corresponds to replacing the exact value of the correlation functions by the value they would obtain if $n(\mathbf{q})$ were held fixed in time and the correlation functions allowed to approach their asymptotic value. The approximation will be valid whenever the relaxation time of $n(\mathbf{q})$ is much greater than the time scale for the change of the correlation functions. This latter is roughly the time necessary for a magnon of average velocity to traverse a distance on the order of the range of the exchange interaction. To see this, consider one of the integrals that results from substituting (14) into (3) and eliminating the momentum delta function:

$$\begin{aligned}
I(\mathbf{q}_1,\ z) &= 2N^{-2}\\
&\times\sum_{\mathbf{q},\mathbf{q}_2}\Gamma(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_1+\mathbf{q},\ \mathbf{q}_2-\mathbf{q})\Gamma(\mathbf{q}_1+\mathbf{q},\ \mathbf{q}_2-\mathbf{q};\ \mathbf{q}_1\mathbf{q}_2)\\
&\quad\times(z+\omega(\mathbf{q}_1+\mathbf{q})-\omega(\mathbf{q}_1)+\omega(\mathbf{q}_2-\mathbf{q})-\omega(\mathbf{q}_2))^{-1}\\
&\quad\times\tilde{n}(\mathbf{q}_2)*\tilde{n}(\mathbf{q}_1)*(1+\tilde{n}(\mathbf{q}_1+\mathbf{q})+\tilde{n}(\mathbf{q}_2-\mathbf{q})).
\end{aligned} \tag{15}$$

If we fix $\mathbf{q}_2$, the integrand is a smoothly varying function of $\mathbf{q}$ over a range of wave vectors on the order of the reciprocal of the range of the exchange interaction, which we denote by $\bar{\mathbf{q}}$. We are ultimately interested in the behavior of $n(\mathbf{q}_1,\ z)$ for $z\cong-i\tau(\mathbf{q}_1)^{-1}$ where $\tau(\mathbf{q}_1)$ is an effective relaxation time for $n(\mathbf{q}_1)$. In this region, replacing the value

$$C(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4z)$$

by its value on the real axis,

$$C(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4 i\epsilon),$$

will be valid if the imaginary part of the factor

$$[-i\tau(q_1)^{-1}+(\omega(\mathbf{q}+\mathbf{q}_1)-\omega(\mathbf{q}-\mathbf{q}_2)-\omega(\mathbf{q}_1)-\omega(\mathbf{q}_2))]$$

behaves essentially as a delta function in the integral. This will be the case as long as the width in wave vectors of this Lorentzian is much smaller than $\bar{\mathbf{q}}$. This is true, on the average over $\mathbf{q}_2$, if $\tau(\mathbf{q}_1)\bar{\mathbf{q}}\bar{V}\gg 1$ where $\bar{V}$ is an average value of $\nabla_{\mathbf{q}_1}\omega(\mathbf{q}_1)-\nabla_{\mathbf{q}_2}\omega(\mathbf{q}_2)$. This is precisely the condition that the collision time, or the time necessary for an average particle to move a distance on the order of the range of the interaction, be much less than the relaxation time. This condition fails for small $\mathbf{q}_1$. However, there is very little contribution to the final integrals from this region, since the phase space available is small. An essentially identical argument forms the basis for our neglect of the initial values of $C(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4)$.

Thus, in so far as the final equation of motion is concerned, we can approximate $C(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4 z)$ by

$$(2S)^{-2}C(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4,\ i\epsilon)=2\delta(\mathbf{q}_1+\mathbf{q}_2-\mathbf{q}_3-\mathbf{q}_4)\left[\frac{P}{\omega(\mathbf{q}_1)+\omega(\mathbf{q}_2)-\omega(\mathbf{q}_3)-\omega(\mathbf{q}_4)}-i\pi\delta(\omega(\mathbf{q}_1)+\omega(\mathbf{q}_2)-\omega(\mathbf{q}_3)-\omega(\mathbf{q}_4))\right]$$
$$\times[\Gamma(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4)\tilde{n}(\mathbf{q}_3)*\tilde{n}(\mathbf{q}_4)*(1+\tilde{n}(\mathbf{q}_1)+\tilde{n}(\mathbf{q}_2))-\Gamma(\mathbf{q}_3\mathbf{q}_4;\ \mathbf{q}_1\mathbf{q}_2)\tilde{n}(\mathbf{q}_1)*\tilde{n}(\mathbf{q}_2)*(1+\tilde{n}(\mathbf{q}_3)+\tilde{n}(\mathbf{q}_4))].\quad(16)$$

The symbol $P$ denotes that the principal value should be used when this term is inserted in an integral.

It should be noted that the real part of $C(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4 i\epsilon)$ is symmetric under the interchange of $\mathbf{q}_1\mathbf{q}_2\leftrightarrow\mathbf{q}_3\mathbf{q}_4$, while the imaginary part is antisymmetric. The sums in equation (3) can be written, when $\mathbf{q}_1=\mathbf{q}_2$, as

$$(2SN)^{-1}\sum_{\mathbf{q}_2'\mathbf{q}_3'\mathbf{q}_4'}\Gamma(\mathbf{q}_1\mathbf{q}_2';\ \mathbf{q}_3'\mathbf{q}_4')[C(\mathbf{q}_3'\mathbf{q}_4';\ \mathbf{q}_1\mathbf{q}_2')-C(\mathbf{q}_1\mathbf{q}_2';\ \mathbf{q}_3'\mathbf{q}_4')]$$

and hence only the antisymmetric part of (15) contributes to the final equation of motion.

We note that although $\Gamma(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4)\neq\Gamma(\mathbf{q}_3\mathbf{q}_4;\ \mathbf{q}_1\mathbf{q}_2)$, the difference is zero on the energy shell for which $\omega(\mathbf{q}_1)+\omega(\mathbf{q}_2)-\omega(\mathbf{q}_3)-\omega(\mathbf{q}_4)=0$. The asymmetry of $\Gamma$ has as a consequence the non-Hermiticity of Dyson's Hamiltonian in terms of boson operators. Making use of the symmetry of $\Gamma$ when multiplied by the energy and momentum delta functions, substituting (16) into (3), and converting back to the time domain, we have finally the equation of motion for $n(\mathbf{q})$, (17), which should be recognized as the quantum-mechanical Boltzmann equation, with the Born approximation for the scattering amplitude appropriate to Dyson's dynamical interaction:

$$(\partial/\partial t)n(\mathbf{q}_1)=-4\pi N^{-2}\sum_{\mathbf{q}_2\mathbf{q}_3\mathbf{q}_4}[\Gamma(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4)]^2\times\delta(\mathbf{q}_1+\mathbf{q}_2-\mathbf{q}_3-\mathbf{q}_4)\delta(\omega(\mathbf{q}_1)+\omega(\mathbf{q}_2)-\omega(\mathbf{q}_3)-\omega(\mathbf{q}_4))\times\{n(\mathbf{q}_1)n(\mathbf{q}_2)[1+n(\mathbf{q}_3)][1+n(\mathbf{q}_4)]-n(\mathbf{q}_3)n(\mathbf{q}_4)[1+n(\mathbf{q}_2)][1+n(\mathbf{q}_1)]\}.\quad(17)$$

It might be argued that Eq. (17) could simply be written down on the basis of Dyson's boson Hamiltonian, and existing derivations of the master equation.[12,13] These derivations make use of the Hermiticity of the Hamiltonian in order to achieve detailed balance, however, and hence are not immediately applicable to the effective boson Hamiltonian, which, as a consequence of the asymmetry of $\Gamma$, is not Hermitian. Furthermore, we feel that our derivation of the Boltzmann equation brings out clearly the part that distinct time scales play in the dynamics. In this respect, our derivation is similar in spirit to the method used by Prigogine[14] and his co-workers, although they base their arguments on a perturbation expansion of the full Liouville operator, rather than a decoupling of the equations of motion for the reduced density matrices, or Wigner functions. Our derivation also permits a straightforward discussion of the significance of kinetic interactions and the interpretation of the Boltzmann equation as a low-density expansion.

The collision term in (17) guarantees that in the evolution of the velocity distribution function, the quantities $\sum_{\mathbf{q}}n(\mathbf{q})$, $\sum_{\mathbf{q}}\mathbf{q}n(\mathbf{q})$, $\sum_{\mathbf{q}}\omega(\mathbf{q})n(\mathbf{q})$ do not change.[15] These quantities can be interpreted as the total number, momentum, and energy of the magnon gas. Except for the total number in the case that $S=\frac{1}{2}$, these are not exact conservation laws for the system, but are a consequence of our approximations. Furthermore, the collision term guarantees that small disturbances in the distribution function relax in a time $\tau(\mathbf{q})$ given by

$$\tau(\mathbf{q}_1)^{-1}=4\pi N^{-2}\sum_{\mathbf{q}_2\mathbf{q}_3\mathbf{q}_4}[\Gamma(\mathbf{q}_1\mathbf{q}_2;\ \mathbf{q}_3\mathbf{q}_4)]^2\times\delta(\mathbf{q}_1+\mathbf{q}_2-\mathbf{q}_3-\mathbf{q}_4)\delta(\omega(\mathbf{q}_1)+\omega(\mathbf{q}_2)-\omega(\mathbf{q}_3)-\omega(\mathbf{q}_4))\times n(\mathbf{q}_2)(1+n(\mathbf{q}_3))(1+n(\mathbf{q}_4)).\quad(18)$$

12 L. Van Hove, Physica **23**, 441 (1957).
13 P. M. Mathews, Physica **32**, 2007 (1966).
14 I. Prigogine, *Nonequilibrium Statistical Mechanics* (Interscience Publishers, Inc., New York, 1962).
15 K. Huang, *Statistical Mechanics* (John Wiley & Sons, Inc., New York, 1963).

The equilibrium distribution can no longer be arbitrary, but must be of the Bose-Einstein form

$$\bar{n}(\mathbf{q})=(\exp\{\beta[\omega(\mathbf{q})+\boldsymbol{\gamma}\cdot\mathbf{q}+\mu]\}-1)$$

if it is to be a stationary solution of (17). $\beta$, $\boldsymbol{\gamma}$, and $\mu$ are in principle arbitrary, but we shall take $\boldsymbol{\gamma}$, $\mu$ to be zero, so that $\bar{n}(\mathbf{q})$ is given by its noninteracting spin-wave value.

Equation (18) is the relaxation time that would be computed from Dyson's dynamical interaction using the Born approximation. We shall make the quadratic approximation to the function $V(\mathbf{q})$, and define an effective magnon mass by the relation

$$\hbar S[V(0)-V(\mathbf{q})]=\hbar^2\mathbf{q}^2/2m.$$

For a simple cubic lattice with nearest-neighbor exchange $m=3\hbar/SV(0)a^2$, where $a$ is the lattice parameter. The relaxation time $\tau(\mathbf{q})$ is then explicitly, to lowest order in $kT$, when $H=0$,

$$\tau(\mathbf{q})^{-1}=[V(0)/6S](kTm/2\pi\hbar^2)^{5/2}a^5(\mathbf{q}a)^3\xi(5/2), \quad (19)$$

where $\xi$ is the Riemann zeta function. The quantity $(2\pi\hbar^2/kTm)^{1/2}$ is the thermal wavelength for the magnons, $\lambda_T$. Equation (19) holds only for wavelengths shorter than the thermal wavelength. At low temperatures, the density of thermally excited magnons, $\bar{n}=S-\langle\langle S^z\rangle\rangle$, is

$$S-\langle\langle S^z\rangle\rangle=\xi(3/2)(a/\lambda_T)^3. \quad (20)$$

There are three distinct changes that occur in the equation of motion for $(1/2S)\langle S^-(\mathbf{q}'+\frac{1}{2}\mathbf{q})S^-(\frac{1}{2}\mathbf{q}-\mathbf{q}')\rangle$ when we allow $\mathbf{q}$ to be nonzero. The most important of these is the appearance of the free drift terms for the magnons, arising from the first term on the right of Eq. (3). There are, in addition, Hartree-Fock terms such as we have already considered, as well as modifications to these terms arising from $C(\mathbf{q}_1\mathbf{q}_2;\,\mathbf{q}_3\mathbf{q}_4)$ because of the fact that the real part of this quantity is no longer completely symmetric in the exchange of $\mathbf{q}_1\mathbf{q}_2\leftrightarrow\mathbf{q}_3\mathbf{q}_4$. And finally there are modifications of the collision term that arise from the fact that the Wigner function varies over the range of the collisions. The full nonlinear equations are thus quite complicated although they can be obtained in a straightforward way by the procedure we have outlined. We are primarily interested in the long-wavelength, linear response function. As we have already seen, the contribution of the Hartree-Fock terms to the linearized response function is negligible for $\mathbf{q}$ small and for all $\omega$, as long as $S-\langle\langle S^z\rangle\rangle\ll S$. We will, therefore, neglect the effect of these terms on the linearized equations of motion, and include only the slight modifications that result from the energy renormalization of the magnons. The corrections arising from the real part of $C(\mathbf{q}_1\mathbf{q}_2;\,\mathbf{q}_3\mathbf{q}_4 i\epsilon)$ are neglected for the same reason.

The corrections to the collision integral arise from the variation of the magnon-distribution function over distances comparable to the range of the exchange interaction, and hence will be quite small for wavelengths long compared to this range. Furthermore, these corrections do not affect the conservation laws to lowest order in $\mathbf{q}$. We will therefore neglect these corrections also.

For the reasons mentioned, it is a good approximation in the long-wavelength limit, to represent the linearized equation of motion for the magnon distribution function by the sum of a drift term and a collision term:

$$(\partial f/\partial t)(\mathbf{q},\mathbf{q}')-(i\mathbf{q}\cdot\hbar\mathbf{q}'/m^*)f(\mathbf{q},\mathbf{q}')=\mathcal{L}_c[f(\mathbf{q},\mathbf{q}')], \quad (21)$$

where $\mathcal{L}_c[f(\mathbf{q},\mathbf{q}')]$ can be obtained by making the replacement

$$n(\mathbf{q}')\leftarrow\bar{n}(\mathbf{q}')+\epsilon f(\mathbf{q},\mathbf{q}')$$

in Eq. (17) and keeping only terms of order $\epsilon$.

Here, $m^*$ is the renormalized magnon mass defined by the relation $\hbar\omega_1(\mathbf{q})=\hbar H+(\hbar^2\mathbf{q}^2/2m^*)$, with $\omega_1(\mathbf{q})$ defined as in Eq. (5), and the quadratic approximation used for $V(\mathbf{q})$.

It is well known[15] that the solutions of (21) in the long-wavelength limit are of the form of linearized local equilibrium solutions. These are eigenfunctions of the operator $\mathcal{L}_c$ with eigenvalue zero, and can be written as

$$f_{\text{LE}}(\mathbf{q},\mathbf{q}',z)=-(1+\bar{n}(\mathbf{q}'))\bar{n}(\mathbf{q}')[\alpha(\mathbf{q},z)+\boldsymbol{\gamma}(\mathbf{q},z)\cdot\mathbf{q}'+\beta(\mathbf{q},z)\omega(\mathbf{q}')]. \quad (22)$$

The quantities $\alpha(\mathbf{q},z)$, $\beta(\mathbf{q},z)$, and $\boldsymbol{\gamma}(\mathbf{q},z)$ determine the local number, momentum, and energy density, and can be evaluated from the moment equations for these quantities. These follow immediately from (21) upon multiplying that equation by $l$, $\mathbf{q}'$, or $\omega(\mathbf{q}')$, and summing over $\mathbf{q}'$. In order that (22) be a good approximation to the exact solution of (21), it is necessary that the wavelength of the disturbance be much larger than the mean free path of the magnons. We define the mean free path as the product of the thermal velocity, $(2kT/m)^{1/2}$, and $\bar{\tau}$, defined as

$$1/\bar{\tau}=N^{-1}\sum_{\mathbf{q}}\tau(\mathbf{q})^{-1}\bar{n}(\mathbf{q})/N^{-1}\sum_{\mathbf{q}}\bar{n}(\mathbf{q}).$$

The average relaxation frequency as defined in this manner is not significantly different from the value one obtains from (19) using the value of $\mathbf{q}$ appropriate to the thermal velocity. We find for $H=0$,

$$\lambda_c/a=0.1S^2(S-\langle\langle S^z\rangle\rangle)^{-7/3}/2.612, \quad (23)$$

where $\lambda_c$ is the mean free path.

The conservation laws, together with (22), lead immediately to soundlike propagation of the magnetization. A more detailed treatment, which includes the effect of the damping of the soundlike mode, may be obtained by replacing the exact collision integral with an approximate expression such as

$$\mathcal{L}_c[f(\mathbf{q},\mathbf{q}',t)] \leftarrow -(1/\bar{\tau})[f(\mathbf{q},\mathbf{q}',t)-f_{\rm LE}(\mathbf{q},\mathbf{q}',t)].$$

The parameters $\alpha$, $\gamma$, and $\beta$ are then determined self-consistently from the moment equations in such a way that the conservation laws are satisfied. This procedure was initially suggested by Bhatnagar, Gross, and Krook[16] and lends itself readily to a perturbation expansion in powers of $\omega\bar{\tau}$. We find that the susceptibility, to first order in $\omega\bar{\tau}$, is

$$\chi(\mathbf{q},z)=\frac{-[\hbar\mathbf{q}^2(S-\langle\langle S^z\rangle\rangle)/m^*](1-iz\bar{\tau})}{z^2-c^2\mathbf{q}^2+iz\Gamma\mathbf{q}^2+i\kappa\mathbf{q}^2c^2\mathbf{q}^2\{z+i\kappa\mathbf{q}^2[1+\hbar(S-\langle\langle S^z\rangle\rangle)/c^2m^*\chi]\}^{-1}}, \tag{24}$$

where

$$c^2=\frac{5}{3}\frac{[\frac{1}{3}N^{-1}\sum_{\mathbf{q}}(\hbar\mathbf{q}/m^*)^2\bar{n}(\mathbf{q})]}{N^{-1}\sum_{\mathbf{q}}\bar{n}(\mathbf{q})}=\frac{5}{3}\frac{K_BT}{m^*}\frac{g_{5/2}(X)}{g_{3/2}(X)},$$

$$g_n(X)=\sum_{K=1}^{\infty}\left(\frac{X}{K}\right)^n,\qquad X=\exp\left(-\frac{gu_BH}{K_BT}\right),$$

$$\Gamma=\tfrac{4}{5}c^2\bar{\tau},$$

$$\kappa=\left(\frac{7}{3}\frac{g_{7/2}(X)}{g_{5/2}(X)}-\frac{5}{3}\frac{g_{5/2}(X)}{g_{3/2}(X)}\right)\frac{K_BT}{m^*}\bar{\tau},$$

$$\chi=(\partial/\partial\hbar H)N^{-1}\sum\bar{n}(\mathbf{q})=-\hbar^{-1}(\partial\langle\langle S^z\rangle\rangle/\partial H). \tag{25}$$

The quantity $\chi$ is just the isothermal susceptibility that would be computed from noninteracting spin-wave theory. For $z\simeq c\mathbf{q}$, $z\bar{\tau}\ll 1$, the last terms in the denominator are negligible, and the sound-wave dispersion relation becomes

$$\omega=c\mathbf{q}(\pm 1-i\tfrac{2}{5}c\mathbf{q}\bar{\tau}).$$

We have assumed that $\bar{n}(\mathbf{q})$ could be neglected at the zone boundaries in calculating (24) and (25).

## V. VALIDITY OF THE BOLTZMANN EQUATION

The existence of a hydrodynamic mode follows in a straightforward way from the Boltzmann equation. The only pathological point about the magnon interaction is the fact that the very low-momentum magnons have arbitrarily long collision times [Eq. (19)], and one might hesitate to treat their relaxation by means of an average collision-time approximation. However, the phase space available to these magnons is vanishingly small, and the principal contribution to the damping of a sound wave comes from those particles having higher than average momentum, so that we do not expect this to be a significant objection.

The hydrodynamic mode will therefore exist, unless the Boltzmann equation does not provide an accurate description of the dynamics over a time period long compared to an average relaxation time. We wish, therefore, to assess the accuracy of Eq. (21).

In the case that $S=\frac{1}{2}$, for which Eq. (2) is exact, the errors in our derivation stem primarily from our decoupling approximation, since we feel that the additional approximations that were made in order to reduce the equations to their final form are justifiable for the reasons mentioned. The decoupling approximation involves only a six-spin correlation function, and leads to terms in the linearized equations of motion that are proportional to $(\bar{n})^2$. (It is easy to show that $\langle\langle S_i^-S_j^+\rangle\rangle\leq\frac{1}{2}-\langle\langle S^z\rangle\rangle$ for all $i$ and $j$.) Thus terms proportional to $(\bar{n})^2$ vanish at least as fast as $(kT)^3$. Even if we had neglected these terms entirely, we would still have arrived at a linearized Boltzmann equation, but with the classical expression for the collision term, since it would not have included the possibility of a particle being in the final state of the scattering process. We expect, therefore, that the corrections arising from inaccuracies in our decoupling approximation vanish at least as fast as $T^3$. This is unfortunately not sufficiently fast to guarantee that the solution (24) is asymptotically exact in the limit that $\omega$, $\mathbf{q}$, $T\to 0$ in such a way that $w\bar{\tau}\ll 1$, and $8\lambda_c\ll 1$. From (19), we see that $\bar{\tau}$ vanishes as $T^4$. Thus, the collision integral could be negligible compared to the terms we have neglected, and the conservation laws upon which we have based the argument invalid. It seems unlikely that this is the case, but we have no proof that it is not.

If one allows for an external field, then the density of magnons and the temperature of the magnon gas can be varied independently. The relaxation frequency depends linearly on the density, for a fixed temperature, while the corrections that we have neglected will vary as $(\bar{n})^2$. The corrections will be asymptotically negligible if we increase the field, but keep the temperature constant. In the high-field limit, the Boltzmann equation goes over to its classical form, and the sound velocity approaches its classical value. Unfortunately, the strength of the pole in the susceptibility depends linearly upon the density also. Thus, although the existence of the sound wave can be guaranteed in the

[16] J. L. Bhatnagar, E. P. Gross, and M. Krook, Phys. Rev. **102**, 593 (1956).

low-density limit, it would be extremely difficult to observe there.

When $S>\frac{1}{2}$, we cannot relate the sum over $\mathbf{q}'$ of $f(\mathbf{q},\mathbf{q}')$ directly to the change in the magnetization, since Eq. (2) no longer holds. We have instead a relation of the form

$$S^z=S-\frac{S^-S^+}{2S}-\frac{S^-S^-S^+S^+}{(2S)^2(2S-1)}+\cdots.$$

We feel that the additional terms have very little effect on our conclusions. They contribute terms to the equation of motion for $f(\mathbf{q},\mathbf{q}')$ that vanish at least as fast as $(\bar{n})^2$. Hence they do not affect the existence of the sound wave in the low-density limit, and can at most modify the temperature and external field at which the hydrodynamic mode becomes ill defined. In view of the fact that we are unable to make any rigorous statements about the equations to an accuracy of better than $T^3$ anyway, it does not seem worthwhile to try to incorporate the higher-spin corrections into the equation of motion. Physically, we would expect the higher-spin values to make the interacting boson model better, in any case, since the kinematic interaction is presumably less effective.

## VI. INTERACTIONS WITH PHONONS

Direct observation of the hydrodynamic modes by means of a transmission experiment would probably be difficult since one would have to avoid exciting magnetostatic modes. Observation by means of inelastic neutron scattering may not be feasible, because of the weakness of the pole in the susceptibility. We therefore investigate the possibility of observing this mode indirectly, by means of its effect on a phonon system.

A magnetoelastic interaction arising from single-ion anisotropy can contain a term such as

$$\mathcal{H}_{\text{int}}=\hbar G\sum_i \epsilon_i(S_i^z)^2,$$

where $\epsilon_i$ is a strain and $G$ a coupling constant. At low temperatures, the dynamical variable $(S_i^z)^2$ can, in the spirit of a spin-wave approximation, be replaced by $(2S-1)S_i^z$. Such a term leads to a direct coupling between the magnon density fluctuations and the phonons. For simplicity, we consider the interaction with a single phonon mode. The complete Hamiltonian for the system is then

$$\sum_i (P_i^2/2M)+\tfrac{1}{2}\sum_{i,j}C_{ij}U_iU_j+\hbar G(2S-1)\sum_i \epsilon_iS_i^z+\mathcal{H}_{\text{Heisenberg}},$$

where $C_{ij}$ are force constants, $U_i$ the displacements of the ions, and $M$ the mass of the ions. The displacements and momenta satisfy the commutation relations

$$[U_i,P_j]=i\hbar\,\delta_{ij}.$$

We wish to calculate the phonon susceptibility

$$\chi_{\text{ph}}(\mathbf{q},\omega)=-i\int_0^\infty e^{i\omega t}\langle\langle[U(\mathbf{q},t),U(-\mathbf{q})]\rangle\rangle.$$

Let

$$C(\mathbf{q})\equiv N^{-1/2}\sum_i \exp(i\mathbf{q}\cdot r_i-r_j)C_{ij}=MC^2\mathbf{q}^2.$$

The equation of motion for $U(\mathbf{q})$ is then

$$-[\partial^2\langle U(\mathbf{q})\rangle/\partial t]=C^2\mathbf{q}^2\langle U(\mathbf{q})\rangle+i\hbar q(G/M)(2S-1)\langle S^z(\mathbf{q})\rangle. \quad (26)$$

This is to be solved with the initial condition

$$\delta\rho(t=0)=-i[U(-\mathbf{q}),\rho_{\text{eq}}],$$

where $\rho_{\text{eq}}$ is the thermal-equilibrium density matrix for the full system. As far as the spin system is concerned, it starts out from a state for which $\langle S^z(\mathbf{q})\rangle=0$ and responds to an effective external field proportional to $\langle U(\mathbf{q})\rangle$. We have therefore, to lowest order in the coupling constant $G$,

$$\langle S^z(\mathbf{q},t)\rangle=i(2S-1)Gq\int_0^t \chi_H(\mathbf{q},t-t')\langle U(t')\rangle dt',$$

where $\chi_H$ is the susceptibility of the spin system. Substituting this into Eq. (26) and taking Laplace transforms, we find

$$\chi_{\text{ph}}(\mathbf{q},z)=[z^2-C^2\mathbf{q}^2-((\hbar G)^2/M)(2S-1)^2\mathbf{q}^2\chi_H(\mathbf{q},z)]^{-1}.$$

The magnetoelastic interaction is quite weak, and a strong interaction can only be expected when the phonon and magnon sound velocities are equal. In this case, we have, using (24),

$$\chi_{\text{ph}}(\mathbf{q},z)=[z^2-C^2\mathbf{q}^2+(\Gamma^2\mathbf{q}^4/z^2-C^2\mathbf{q}^2)]^{-1},$$

where $\Gamma=(2S-1)\hbar G[(S-\langle\langle S^z\rangle\rangle)/mM]^{1/2}$. For small values of $\Gamma/C$, $\chi_{\text{ph}}$ has poles at

$$\omega=\pm Cq-\tfrac{1}{2}i(\Gamma/C)q.$$

If we express the sound velocity in terms of a force constant, $C^2=k/M$, then the acoustic attenuation coefficient $\alpha$, defined as $\Gamma/2C^2$ can be written in terms of a magnetoelastic interaction parameter $\gamma=\hbar G/k$:

$$\alpha=[(2S-1)/2]\gamma(M(S-\langle\langle S^z\rangle\rangle)/m)^{1/2}. \quad (27)$$

If the additional attenuation of the phonons due to their interaction with the magnon sound wave is to be observed, $\alpha \gtrsim 10^{-3}$. In terms of the Curie temperature, the magnon mass is roughly $m=2(S+1)\hbar^2/KTca^2$. With $S=1$, $a=4$ Å, a Curie temperature of 300° corresponds to a magnon mass of approximately 0.04 amu, and a maximum sound velocity, if (25) were valid at the Curie temperature (which it is not) of approximately $10^6$ cm/sec. There would thus be an ample range of temperatures at which the magnon sound velocity was in the phonon range. If the ions are massive, the square root can easily be on the order of 25 for $S-\langle\langle S^z\rangle\rangle \simeq 0.2$, so that the values of $\gamma$ that are required would be on the order of $4\times10^{-5}$. This is a reasonable order of magnitude for a magnetostriction constant.

## VII. CONCLUSION

If collisions between magnons are neglected, the longitudinal susceptibility of the Heisenberg spin system at low temperatures is essentially that which would be derived from the noninteracting spin-wave model. As long as $S-\langle\langle S^z\rangle\rangle \ll 2S$, the "average field" effects arising from the interactions are negligible, and do not produce any collective motions. When the effect of collisions is taken into account, it is found that the magnon velocity distribution satisfies the quantum-mechanical Boltzmann equation. Since the "average field" effects are negligible, we have argued that the Fourier transform of the distribution of magnons in phase space, defined as

$$(1/2S)\langle S^-(\tfrac{1}{2}\mathbf{q}+\mathbf{q}')S^+(\tfrac{1}{2}\mathbf{q}-\mathbf{q}')\rangle,$$

satisfies an equation of motion that is adequately approximated, in the long-wavelength limit, by the sum of a free drift term and the Boltzmann collision term. The susceptibility may be obtained from the linearized hydrodynamic equations that follow immediately from the form of the Boltzmann collision term. We conclude that the susceptibility has a pole at $\omega=\pm C\mathbf{q}$, where $C$ is the velocity of magnon sound, given by (25). The decoupling procedure we used to obtain the Boltzmann equation is accurate up to terms of order $(S-\langle\langle S^z\rangle\rangle)^2$. Although this is not sufficient to justify the results we have obtained as an asymptotic expansion in the temperature $T$, it is sufficient to justify them, in the presence of an external field, in terms of an asymptotic expansion in the density of thermally excited magnons, in which case the Boltzmann equation assumes its classical form. The interaction of the magnon sound wave with a model phonon system through single-ion anisotropy was considered. We conclude that the interaction could produce an observable increase in the phonon lifetime when the temperature is adjusted so that the magnon sound velocity coincides with the phonon velocity.


## ACKNOWLEDGMENTS

I would like to thank Dr. Marshall Sparks and Dr. Robert M. White for their advice and encouragement, and the Center for Materials Research at Stanford for their support during the early phases of this work.


## APPENDIX

The scale of changes in $\bar{n}(\mathbf{q})$ is given by the reciprocal of the thermal wavelength. Hence, when the wavelength of the external disturbance is much greater than the thermal wavelength, we can replace the finite differences in the denominator of (8) by differentials. Upon converting the sums to integrals we obtain for the denominator in (8), along the real axis,

$$1+\frac{V}{(2\pi)^3}\int\frac{d^3\mathbf{q}'\mathbf{q}\cdot\nabla_{\mathbf{q}}'[(V(0)-V(\mathbf{q}'))n(\mathbf{q}')]}{\omega+i\epsilon+\mathbf{q}\cdot\nabla_{\mathbf{q}}'\omega(\mathbf{q}')}. \quad \text{(A1)}$$

Using the quadratic approximation, and defining $W=m\omega/\hbar|\mathbf{q}|$, this becomes

$$1+(1/2S)\int_{-\infty}^{\infty}\frac{g(\mu)d\mu}{W+\mu+i\epsilon},$$

where

$$g(\mu)=\frac{V}{(2\pi)^3}\int d^3\mathbf{q}\,\delta\left(\mu-\frac{\mathbf{q}\cdot\mathbf{q}'}{|\mathbf{q}|}\right)\frac{\mathbf{q}}{|\mathbf{q}|}\cdot\nabla_{\mathbf{q}'}(\mathbf{q}'^2n(\mathbf{q}')).$$

Replacing the exact zone boundary by a sphere, and doing the angular integrations, gives

$$g(\mu)=\frac{V}{(2\pi)^2}\int_{|\mu|}^{\mathbf{q}'_{\max}}2\mu\mathbf{q}'\frac{\partial}{\partial(\mathbf{q}')^2}(\mathbf{q}'^2n(\mathbf{q}'))d\mathbf{q}'.$$

We will neglect the contribution from the zone boundary and so obtain the result that (A1) can be written

$$1-\frac{V}{(2\pi)^2}(2S)^{-1}\int_{-\infty}^{\infty}\frac{\mu^3n(\mu)d\mu}{W+\mu+i\epsilon}. \quad \text{(A2)}$$

The real part of the integral is a maximum at $W=0$, where it has the value $S-\langle\langle S^z\rangle\rangle/2S$. In terms of the variable $x=m^{1/2}\omega/q(2kT)^{1/2}$, the imaginary part may be written

$$(2S)^{-1}(V/4\pi)(2kTm/\hbar^2)^{3/2}x^3(e^{x^2}-1)^{-1}. \quad \text{(A3)}$$

The maximum of the function $x^3(e^{x^2}-1)^{-1}$ is approximately 0.6. Using (20), (A3) can be written as

$$(4\pi)^{1/2}[S-\langle\langle S^z\rangle\rangle/2S]x^3(e^{x^2}-1)\leq S^{-1}(S-\langle\langle S^z\rangle\rangle).$$

Thus both the real and imaginary parts of the integral are uniformly small, for all $\omega$, when $S-\langle\langle S^z\rangle\rangle \ll S$.