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Hydrodynamics of Heisenberg Ferromagnets

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A microscopic derivation is given of hydrodynamic equations and response functions for uniaxial and isotropic exchange Heisenberg ferromagnets. The method is based on the conservation laws and makes no use of a quasiparticle picture. At low temperatures, the hydrodynamic equations involve the z component of the magnetization, the local temperature, and the approximately conserved momentum. In addition to a diffusive mode, there is a propagating mode, the second magnon, which is strongly coupled to the z component of the magnetization and may be observed by neutron scattering, and, in transparent ferromagnets, by Brillouin scattering (e.g., in Eu compounds and $CrBr_3$). The strength of this mode depends on the magnetic field and anisotropy. For zero external field and no anisotropy, the longitudinal susceptibility diverges for wave number $q\rightarrow 0$. The velocities and diffusion constants in the hydrodynamic equations become q dependent. Then, also, the transverse spin components perform low-frequency oscillations. For higher temperatures, the momentum is no longer conserved, and the hydrodynamic equations reduce to two coupled diffusion equations for the local temperature and magnetization. For $T>T_c$ and zero external field, the spin density and energy obey uncoupled diffusion equations.

I. INTRODUCTION

At finite temperature, the low-frequency response and excitations of a many-particle system are described by hydrodynamic equations. This paper is devoted to a quantum-mechanical derivation of hydrodynamics for uniaxial and isotropic Heisenberg ferromagnets. The method employed is closely related to the general theory of Brownian motion and transport by Mori.[1,2] It is based on the microscopic conservation laws and the fact that in the hydrodynamic regime the nonconserved operators relax much faster than the conserved quantities. Since no explicit use is made of the existence of quasiparticles, the derivation also holds in a temperature region where the quasiparticle concept does not apply. For $T>T_c$ and zero external field, one finds a set of uncoupled diffusion equations for the spin density and the energy density. For $T_D \lesssim T < T_c$ (where T_D is the Debye temperature of the magnons), one finds two coupled diffusion equations for the local temperature and the z component of magnetization. (z is the direction of spontaneous magnetization.) For $T \ll T_D$, momentum is conserved approximately which allows for the possibility of an oscillatory motion of momentum, the z component of magnetization, and the local temperature. In analogy to He II and lattice dynamics, this mode is called second magnon.[3] Special attention is given to the conditions for the observability of this mode. Since neutron scattering and, in transparent ferromagnets, Brillouin scattering are most appropriate tools to detect these modes, expressions for the dynamic form factor are derived. Whereas in He II and in lattice dynamics, second sound is mainly a temperature wave [coupled to the density by the small ratio $(c_P - c_V)/c_P$], the second magnon is for finite magnetic field primarily a magnetization wave and therefore has more similarity to a density wave of a real gas. In the magnetic case, one is in the fortunate position of having an extra experimental degree of freedom (the external field) by means of which one may tune the ratio of the contributions of the propagating and the diffusive modes in the dynamic form factor of the magnetization. Since increasing the magnetic field also reduces the value of the static longitudinal susceptibility which enters as a factor in the neutron cross section, an upper limit is set to the applied field. With increasing temperature the propagating mode gets heavily damped and finally one enters the diffusive regime.

In an isotropic ferromagnet in zero external field the transverse components of the spin density also perform low-frequency oscillations. Moreover the longitudinal susceptibility diverges and the coefficients in the hydrodynamic equations are q dependent. (The divergence of the homogeneous longitudinal susceptibility for $H\rightarrow 0$, which usually is derived in spin-wave approximation, follows rigorously from Dyson's theory.[4])

We omit dipole forces and interaction with phonons, which in the temperature range in which we are interested, are indeed negligible. For temperatures where these interactions become important, the hydrodynamic region is restricted to extremely small values of the wave number q.

In Sec. II, we present the general method for deriving hydrodynamic equations.

In Sec. III, we derive the hydrodynamics for the Heisenberg model; in Sec. III B for temperatures where the momentum is not conserved; in Sec. III C for the case of (approximately) conserved momentum.

In Sec. IV, we discuss the solutions of the hydrodynamic equations for finite external field or anisotropy so that the thermodynamic limit ($q \to 0$) in the evaluation of the static susceptibilities may be taken. The transition from the propagating to the diffusive regime is described. The coefficients of the hydrodynamic equations assume a particularly simple form if one generalizes thermodynamics so as to include the volume as a thermodynamic variable.

In Sec. V, we discuss the case of an isotropic ferromagnet in zero external field. Besides the hydrodynamic equations involving the local temperature, magnetization, and momentum, the oscillations of the transverse spin components are also studied.

In Sec. VI, the range of validity of the theory is ascertained. Estimates for the various collision times and criteria for the observability of the second magnon mode by neutron and Brillouin scattering are derived. Special attention is given to EuO and $CrBr_3$.

In Appendix A, we collect a variety of thermodynamic relations. In Appendix B, we derive the changes resulting from the addition of an anisotropic exchange term to the Hamiltonian. In Appendix C, the coefficients of the hydrodynamic equations are calculated in the lowest-order magnon model and in a collision-time approximation.

II. GENERAL DERIVATION OF HYDRODYNAMIC EQUATIONS

In this section, we describe a general quantum-mechanical derivation of hydrodynamic equations based on conservation laws. The method includes the case of approximately conserved quantities such as quasimomentum in a lattice.

The response of a system to perturbations of long wavelength λ and low frequency ω is determined by hydrodynamic equations. In this regime, the quasiparticles undergo frequent collisions within a distance λ and period ω^{-1}. It is important to realize that in this region the variables describing the system separate clearly into two classes whose relaxation times are of completely different orders. The relaxation time of the first group – the densities of conserved operators – is very large, being proportional to the wavelength. On the other hand, the nonconserved operators decay very rapidly to local equilibrium because of the frequent collisions of the quasiparticles. Consequently, the nonequilibrium state can be characterized completely by specifying the spatial and temporal variation of the conserved densities. The time change of these densities $\rho(\vec{x})$ is governed by the conservation laws:

$$\dot{\rho}(\vec{x}) = -\operatorname{div}\vec{j}(\vec{x}) \quad . \tag{2.1}$$

The currents $\vec{j}$ themselves decompose into a part proportional to the densities $\rho(\vec{x})$ and into a nonconserved part. The first will give rise to cooperative oscillatory motions of the conserved densities whereas the latter (like a random force on Brownian particles) will produce damping.[1,2]

Although all the various correlation functions display the hydrodynamic singularities, it seems that Kubo's relaxation function[5] is most appropriate for a microscopic derivation of hydrodynamic equations. In order to demonstrate this, let us imagine that the system is disturbed by adiabatically switching on an external force $K^j(x)$, which varies slowly in space:[6]

$$\delta\mathcal{H}(t) = -\int d^3x\, X^j(\vec{x}, t) K^j(\vec{x}) e^{\epsilon t}\theta(-t) \quad . \tag{2.2}$$

Here $X^j(\vec{x}, t)$ is some operator of the system, whose Fourier transform is denoted by

$$X^j_{\vec{q}}(t) = \int d^3x\, e^{-i\vec{q}\cdot\vec{x}} X^j(\vec{x}, t) \quad .$$

At time $t = 0$ the disturbance is turned off and we observe the system relaxing to equilibrium. The relaxation of the operator X^i is given by

$$\delta\langle X^i_{\vec{q}}(t)\rangle = \Phi^{ij}(\vec{q}, t) K^j(\vec{q}) \quad \text{for} \quad t > 0 \quad . \tag{2.3}$$

Here $\Phi^{ij}(\vec{q}, t)$ is Kubo's relaxation function[5,7]:

$$\Phi^{ij}(\vec{q}, t) = i\int_t^\infty dt'\, e^{-\epsilon t'} \langle [X^i_{\vec{q}}(t'), X^j_{\vec{q}}(0)^\dagger]\rangle \quad . \tag{2.4}$$

Its one-sided Fourier transform is related to the more commonly used response function

$$\chi^{ij}(\vec{q}, \omega) = \int_{-\infty}^\infty dt\, e^{i\omega t} i\theta(t) \langle [X^i_{\vec{q}}(t), X^j_{\vec{q}}(0)^\dagger]\rangle \quad , \tag{2.5}$$

by $\Phi^{ij}(\vec{q}, z) \equiv \int_0^\infty dt \Phi^{ij}(\vec{q}, t) e^{izt}$

$$= (1/iz)[\chi^{ij}(\vec{q}, z) - \chi^{ij}(\vec{q}, 0)], \quad \operatorname{Im} z > 0 \quad . \tag{2.6}$$

If the disturbance (2.2) couples to a conserved density, the relaxation is alternatively described macroscopically by hydyodynamic equations. This relation between the relaxation function $\Phi(\vec{q}, t)$ and the initial-value problem of the hydrodynamic equations has been used by Kadanoff and Martin[6] to determine $\Phi(\vec{q}, t)$ and then the dynamic susceptibilities from the known hydrodynamic equations of a liquid. Conversely, if one has to derive the hydrodynamic equations microscopically, it is natural to investigate Kubo's relaxation function Φ. In the following it will be convenient to measure all susceptibilities on a unique scale. This is achieved by orthonormalizing our complete set of operators:

$$\langle X^i_{\vec{q}}, X^j_{\vec{q}} \rangle = \delta^{ij} \quad . \tag{2.7}$$

The scalar product of any two operators is taken to be the static susceptibility

$$\langle A_{\vec{k}}, B_{\vec{k}} \rangle \equiv \chi^{AB}(\vec{k}, \omega = 0) \quad . \tag{2.8}$$

As already stressed we group the operators $X^i_{\vec{q}}$ into two sets: the conserved and the nonconserved operators. If we want to make a distinction between them we shall label the former by $c, c', \ldots,$ and the latter by $n, n' \ldots$. The separation in slowly and rapidly decaying operators has been used by Wegner[2] in a similar derivation.

The time derivative of the operators given by Heisenberg's equation may be expanded in the following manner:

$$\begin{aligned} \dot{X}^c_{\vec{q}} &= -iC^{cc'}(\vec{q})X^{c'}_{\vec{q}} - iC^{cn}(\vec{q})X^n_{\vec{q}} \quad , \\ \dot{X}^n_{\vec{q}} &= -iC^{nc}(\vec{q})X^c_{\vec{q}} - iD^{nn'}(\vec{q})X^{n'}_{\vec{q}} \quad . \end{aligned} \tag{2.9}$$

The coefficients obey the symmetry relations

$$C^{cc'*} = C^{c'c}, \quad C^{nc*} = C^{cn}, \quad D^{nn'*} = D^{n'n} \; . \tag{2.10}$$

Summation over repeated indexes is always implied. For an exactly conserved quantity the expansion coefficients $C^{cc'}(\vec{q})$ and $C^{cn}(\vec{q})$ are proportional to q. In a lattice the momentum is only approximately conserved [see Eq. (3.37)]. The time derivative of the momentum $P^\alpha_{\vec{q}}$ equals the sum of the divergence of the momentum flux tensor and a vector $u^\alpha_{\vec{q}}$ which is different from zero only for umklapp processes. The scalar product of $u^\alpha_{\vec{q}}$ and any other operator is therefore proportional to the small wave number $\kappa = a^{-1}e^{-T_D/T}$ for low temperatures, where T_D is the Debye temperature. For low T we shall include the quasimomentum $P^\alpha_{\vec{q}}$ in the set of conserved operators. Consequently, the general form of the equation of motion for an operator $X^c_{\vec{q}}$ is

$$\begin{aligned} \dot{X}^c_{\vec{q}} = -iq^\alpha j^{c\alpha}_{\vec{q}} - iu^c_{\vec{q}} &= -iq^\alpha m^{cc'}_\alpha(\vec{q})X^{c'}_{\vec{q}} \\ &\quad - iq^\alpha m^{cn}_\alpha(\vec{q})X^n_{\vec{q}} - i\kappa r^{cc'}(\vec{q})X^{c'}_{\vec{q}} - i\kappa r^{cn}(\vec{q})X^n_{\vec{q}} \, . \end{aligned} \tag{2.11}$$

From Eq. (2.11), it is easy to see that the matrix elements $C^{cn}(\vec{q})$ and $C^{cc'}(\vec{q})$ are small because of their linear dependence on either q^α or κ. The matrix $D(q)$, on the other hand, stays finite if q and κ go to zero. The analysis in Sec. III shows that $r^{cc'}$ is zero, and we shall therefore omit this coefficient from now on. From (2.8) one readily derives the equation of motion for Φ:

$$\begin{aligned} \omega\Phi^{cc'}(\vec{q}, \omega) = C^{cc''}(\vec{q})\Phi^{c''c'}(\vec{q}\,, \omega) & \\ + C^{cn}(\vec{q})\Phi^{nc'}(\vec{q}, \omega) + i\delta^{cc'} \quad , & \end{aligned} \tag{2.12a}$$

$$\begin{aligned} \omega\Phi^{nc}(\vec{q}, \omega) = C^{nc'}(\vec{q})\Phi^{c'c}(\vec{q}, \omega) & \\ + D^{nn'}(\vec{q})\Phi^{n'c}(\vec{q}, \omega) \quad , & \end{aligned} \tag{2.12b}$$

$$\begin{aligned} \omega\Phi^{nn'}(\vec{q}, \omega) = C^{nc}(\vec{q})\Phi^{cn'}(\vec{q}, \omega) & \\ + D^{nn''}(\vec{q})\Phi^{n''n'}(\vec{q}, \omega) + i\delta^{nn''} , & \end{aligned} \tag{2.12c}$$

$$\begin{aligned} \omega\Phi^{cn}(\vec{q}, \omega) = C^{cc'}(\vec{q})\Phi^{c'n}(\vec{q}, \omega) & \\ + C^{cn'}(\vec{q})\Phi^{n'n}(\vec{q}, \omega) \; . & \end{aligned} \tag{2.12d}$$

Our aim is to derive a set of equations containing $\Phi^{cc'}$ only. We may rewrite (2.12b) and (2.12c):

$$\Phi^{nc}(\vec{q}, \omega) = \left(\frac{1}{\omega 1 - D(\vec{q})}\right)_{nn'} C^{n'c'}(\vec{q})\Phi^{c'c}(\vec{q}, \omega) \quad , \tag{2.12b$'$}$$

$$\begin{aligned} \Phi^{nn'}(\vec{q}, \omega) = \left(\frac{1}{\omega 1 - D(\vec{q})}\right)_{nn''} C^{n''c}(\vec{q})\Phi^{cn'}(\vec{q}, \omega) & \\ + i\left(\frac{1}{\omega 1 - D(\vec{q})}\right)_{nn'} \quad . & \end{aligned} \tag{2.12c$'$}$$

Since inserting (2.12b′) in (2.12a) results in $(\omega 1 - D)^{-1}$ being multiplied by a second-order term, we need only consider Eq. (2.12*c*′) in the limit of small q and κ. Use of (2.12d) for $\Phi^{cn}(\vec{q}, \omega)$ shows that the first term on the right-hand side of (2.12c′) may be neglected. More precisely, we may write

$$\lim_{\omega \to 0} \lim_{q, \kappa \to 0} \left(\frac{1}{\omega 1 - D(\vec{q})}\right)_{nn'} = -i \lim_{\omega \to 0} \lim_{q, \kappa \to 0} \Phi^{nn'}(\vec{q}, \omega) \; . \tag{2.12c$''$}$$

Because of its repeated occurrence we shall denote $\lim_{\omega \to 0} \lim_{q, \kappa \to 0}$ by Lim. Successive use of (2.12b′) and (2.12c″) in (2.12a) leads to[8]

$$\begin{aligned} [\omega\delta^{cc''} - C^{cc''}(\vec{q}) + iC^{cn}(\vec{q})C^{n'c''}(\vec{q}) & \\ \times \mathrm{Lim}\Phi^{nn'}(\vec{q}, \omega)]\Phi^{c''c'}(\vec{q}, \omega) = i\delta^{cc'} \quad . & \end{aligned} \tag{2.13}$$

This is the desired hydrodynamic equation, which by use of Eq. (2.11) can be written

$$\begin{aligned} [\omega\delta^{cc''} - q^\alpha m^{cc''}(\vec{q}) + i\Gamma^{cc''}(\vec{q})] & \\ \times \Phi^{c''c'}(\vec{q}, \omega) = i\delta^{cc'} \quad . & \end{aligned} \tag{2.14}$$

The relaxation coefficient $\Gamma^{cc''}(\vec{q})$ is of second order in q and κ, and expressible by a Kubo formula

$$\begin{aligned} \Gamma^{cc'}(\vec{q}) &= C^{cn}(\vec{q})C^{c''n'*}(\vec{q}) \\ &\quad \times \mathrm{Lim}\tfrac{1}{2}\beta\int_{-\infty}^{\infty} dt\, e^{i\omega t}\langle \tfrac{1}{2}\{X^n_{\vec{q}}(t), X^{n'}_{\vec{q}}(0)^\dagger\}\rangle_c \\ &= q^\alpha q^\beta\, \mathrm{Lim}\tfrac{1}{2}\beta\int_{-\infty}^{\infty} dt\, e^{i\omega t}\langle \tfrac{1}{2}\{\tilde{j}^{c\alpha}_{\vec{q}}(t), \tilde{j}^{c'\beta}_{\vec{q}}(0)^\dagger\}\rangle_c \\ &\quad + q^\alpha\, 2\,\mathrm{Re}\,\mathrm{Lim}\tfrac{1}{2}\beta\int_{-\infty}^{\infty} dt\, e^{i\omega t}\langle \tfrac{1}{2}\{\tilde{j}^{c\alpha}_{\vec{q}}(t), u^{c'}_{\vec{q}}(0)^\dagger\}\rangle_c \\ &\quad + \mathrm{Lim}\tfrac{1}{2}\beta\int_{-\infty}^{\infty} dt\, e^{i\omega t}\langle \tfrac{1}{2}\{u^c_{\vec{q}}(t), u^{c'}_{\vec{q}}(0)^\dagger\}\rangle_c \, . \end{aligned} \tag{2.15}$$

$\{\;,\;\}$ is the anticommutator. We have denoted the nonconserved part of the current $j^{c}_{\vec{q}}$ by

$$\tilde{j}^{c\alpha}_{\vec{q}} = m^{cn}_{\alpha} X^{n}_{\vec{q}} \quad . \tag{2.16}$$

We want to emphasize that the second equality in (2.15) has a symbolic character: the limit $q \to 0$, $\kappa \to 0$ is not to be applied to the q dependence in $C^{cn}(q)$ and $C^{n''c'}(q)$, but only to the Kubo formulas containing the normalized currents. If the susceptibilities are not singular for $q \to 0$, this difference is irrelevant up to $O(q^2)$. In Eq. (2.15), we have used the relation[7]

$$\mathrm{Lim}\,\Phi^{ij}(\vec{q}, \omega) = \mathrm{Lim}\,\tfrac{1}{2}\beta \int_{-\infty}^{\infty} dt\, e^{i\omega t} \langle \tfrac{1}{2}\{X^{i}_{\vec{q}}(t), X^{j}_{\vec{q}}(0)^{\dagger}\}\rangle_c \;,$$

where the cumulant is defined by

$$\langle\{X^{i}_{\vec{q}}(t), X^{j}_{\vec{q}}(0)^{\dagger}\}\rangle_c$$

$$\equiv \langle\{X^{i}_{\vec{q}}(t) - \langle X^{i}_{\vec{q}}(t)\rangle, X^{j}_{\vec{q}}(0)^{\dagger} - \langle X^{j}_{\vec{q}}(0)^{\dagger}\rangle\}\rangle .$$

First, Eq. (2.14) gives the long-wavelength behavior of the relaxation function $\Phi^{cc'}(\vec{q}, \omega)$ and in turn all the other Green's functions:

$$\begin{aligned} &\Phi^{cc'}(\vec{q}, \omega) = i\,[\omega 1 - q^{\alpha} m_{\alpha} + i\Gamma\,]^{-1}_{cc'} \;, \\ \text{and}\quad &\chi''^{cc'}(\vec{q}, \omega) = \omega\, \mathrm{Re}\Phi^{cc'}(\vec{q}, \omega) \;, \end{aligned} \tag{2.17}$$

where χ'' is the imaginary part of χ. Second, we can immediately infer the structure of the classical hydrodynamic equations

$$\frac{d}{dt}\delta X^{c}_{\vec{q}}(t) = [\,-iq^{\alpha} m^{cc''}_{\alpha}(\vec{q}) - \Gamma^{cc''}(\vec{q})\,]\delta X^{c''}_{\vec{q}} \tag{2.18}$$

for the nonequilibrium averages of $X^{c}_{\vec{q}}$,

$$\delta X^{c}_{\vec{q}}(t) = \langle X^{c}_{\vec{q}}(t)\rangle_{\text{Nonequ.}} - \langle X^{c}_{\vec{q}}(t)\rangle \quad . \tag{2.19}$$

The hydrodynamic Eq. (2.18) is constructed such that the relaxation derived from them is identical with the microscopic result (2.3).

III. HYDRODYNAMICS OF HEISENBERG MODEL

In this section, we shall apply the method of Sec. II to derive the hydrodynamic equations for a Heisenberg ferromagnet. In Sec. III A, we give the necessary definitions, in Sec. III B we derive the hydrodynamics for the case where the momentum is not conserved, and in Sec. III C for the case where $\vec{P}_{\vec{q}}$ is conserved. In order to keep the derivation as short and as clear as possible, we shall in this section consider only an isotropic exchange Hamiltonian with external magnetic field, the modification to the results for the anisotropic case being given in Appendix B. In our units Planck's constant and Boltzmann's constant are equal to 1.

A. Heisenberg Model

The Hamiltonian of the system is given by

$$\mathcal{H} = \mathcal{H}^{e} - g\mu_B H \sum_{\vec{l}} S^{z}_{\vec{l}} \quad , \tag{3.1}$$

where $\mathcal{H}^{e}$ is the exchange Hamiltonian

$$\mathcal{H}^{e} = -\frac{1}{2}\sum_{\vec{l}\,\vec{l}'} J(\vec{l} - \vec{l}\,')(S^{+}_{\vec{l}} S^{-}_{\vec{l}'} + S^{z}_{\vec{l}} S^{z}_{\vec{l}'}) . \tag{3.2}$$

The summation runs over all sites $\vec{l}$ of the magnetic lattice. In Eq. (3.2), $\vec{S}_{\vec{l}}$ is the spin operator (in the Heisenberg representation) for the spin localized at site $\vec{l}$ of the magnetic lattice. The time dependence will not be written explicitly. As usual we have introduced

$$S^{\pm}_{\vec{l}} = S^{x}_{\vec{l}} \pm i S^{y}_{\vec{l}} \quad . \tag{3.3}$$

The exchange interaction between the spin at site $\vec{l}$ and the spin at site $\vec{l}\,'$ is denoted by $J(\vec{l} - \vec{l}\,')$ with $J(0) = 0$. We assume that the magnetic lattice has inversion symmetry. The external field points in the z direction; g is the spectroscopic splitting factor, and μ_B is the Bohr magneton. The equal-time commutation relations for the spin operators are

$$\begin{aligned} &[S^{+}_{\vec{l}}, S^{-}_{\vec{l}'}] = 2\delta_{\vec{l},\vec{l}'} S^{z}_{\vec{l}}, \quad [S^{z}_{\vec{l}}, S^{\pm}_{\vec{l}'}] = \pm\,\delta_{\vec{l}\,\vec{l}'} S^{\pm}_{\vec{l}'} \\ &[S^{\pm}_{\vec{l}}, S^{\pm}_{\vec{l}'}] = [S^{z}_{\vec{l}}, S^{z}_{\vec{l}'}] = 0 \quad . \end{aligned} \tag{3.4}$$

We also note that $\vec{S}^{2}_{\vec{l}} = S(S+1)$ for a spin of magnitude S. Later we shall also need the equations of motion for the spin operators

$$\begin{aligned} &\dot{S}^{\pm}_{\vec{l}} = \pm i \textstyle\sum_{\vec{l}} J(\vec{l} - \vec{l}') (S^{z}_{\vec{l}} S^{\pm}_{\vec{l}'} - S^{z}_{\vec{l}'} S^{\pm}_{\vec{l}}) \mp i g\mu_B H S^{\pm}_{\vec{l}} \;, \\ &\dot{S}^{z}_{\vec{l}} = -\textstyle\sum_{\vec{l}'} J(\vec{l} - \vec{l}\,')(S^{x}_{\vec{l}'} S^{y}_{\vec{l}} - S^{y}_{\vec{l}'} S^{x}_{\vec{l}}) \;\; . \end{aligned} \tag{3.5}$$

B. Hydrodynamics without Momentum Conservation

For high temperatures such that umklapp processes[9] are important, the only conserved operators are the z component of the magnetization and the exchange energy. Their Fourier-transformed densities are

$$M_{\vec{q}} = g\mu_B S^{z}_{\vec{q}} = g\mu_B \sum_{\vec{l}} e^{-i\vec{q}\cdot\vec{x}_{\vec{l}}} S^{z}_{\vec{l}} \tag{3.6}$$

and

$$\mathcal{H}^{e}_{\vec{q}} = -\sum_{l,l'} e^{-i\vec{q}\cdot\vec{x}_{\vec{l}}} \tfrac{1}{2} J(\vec{l} - \vec{l}\,')\, \vec{S}_{\vec{l}}\cdot\vec{S}_{\vec{l}'} \quad . \tag{3.7}$$

If $H = 0$ also $\sum_{\vec{l}} S^{+}_{\vec{l}}$ and $\sum_{\vec{l}} S^{-}_{\vec{l}}$ are conserved. However, for symmetry reasons these operators do not couple to the hydrodynamic equations involving $M_{\vec{q}}$ and $\mathcal{H}^{e}_{\vec{q}}$, and therefore we shall postpone the discussion of S^{+} and S^{-} until Sec. V. From Eqs. (3.5) we find the conservation laws

$$\dot{M}_{\vec{q}} = -iq^{\alpha} j^{M\alpha}_{\vec{q}} \quad , \tag{3.8}$$

$$\dot{\mathcal{H}}^{e}_{\vec{q}} = -iq^{\alpha} j^{e\alpha}_{\vec{q}} \quad , \tag{3.9}$$

where the magnetization and energy currents are given by

$$j^{M\alpha}_{\vec{q}} = g\mu_B \sum_{\vec{l},\vec{l}'} \exp[-\tfrac{1}{2} i\vec{q}\cdot(\vec{x}_{\vec{l}} + \vec{x}_{\vec{l}'})]$$

$$\times \tfrac{1}{2}(x_{\vec{l}} - x_{\vec{l}'})^{\alpha}(S^x_{\vec{l}} S^y_{\vec{l}'} - S^y_{\vec{l}} S^x_{\vec{l}'}) J(\vec{l} - \vec{l}') \qquad (3.10)$$

and

$$j^{e\alpha}_{\vec{q}} = \tfrac{1}{4} \sum_{\vec{l},\vec{l}',\vec{l}''} \exp[\tfrac{1}{2} i\vec{q}\cdot(\vec{x}_{\vec{l}} + \vec{x}_{\vec{l}''})]$$

$$\times (x_{\vec{l}} - x_{\vec{l}''})^{\alpha} J(\vec{l} - \vec{l}') J(\vec{l}' - \vec{l}'') S_{\vec{l}} \cdot S_{\vec{l}'} \times S_{\vec{l}''} \ . \qquad (3.11)$$

Following the program of Sec. II we construct the orthonormal operators:

$$X^3_{\vec{q}} \equiv \langle M_{\vec{q}}, M_{\vec{q}}\rangle^{-1/2} M_{\vec{q}} \ , \qquad (3.12)$$

$$X^4_{\vec{q}} \equiv \frac{1}{d(\vec{q})} (\langle M_{\vec{q}}, M_{\vec{q}}\rangle \mathcal{H}^e_{\vec{q}} - \langle \mathcal{H}^e_{\vec{q}}, M_{\vec{q}}\rangle M_{\vec{q}}) \ . \qquad (3.13)$$

(We reserve labels 1 and 2 for S^+ and S^-, which will be treated later.) The normalization factor $d(\vec{q})$ is given by

$$d(\vec{q}) = [(\langle M_{\vec{q}}, M_{\vec{q}}\rangle \langle \mathcal{H}^e_{\vec{q}}, \mathcal{H}^e_{\vec{q}}\rangle$$

$$- |\langle M_{\vec{q}}, \mathcal{H}^e_{\vec{q}}\rangle|^2) \langle M_{\vec{q}}, M_{\vec{q}}\rangle]^{1/2} \ . \qquad (3.14)$$

We shall see (Appendix A) that in the thermodynamic ($q \to 0$) limit $X^4_{\vec{q}}$ is the operator whose nonequilibrium expectation value is proportional to the local temperature.

For symmetry reasons, the projections of the currents $j^{M\alpha}_{\vec{q}}$ and $j^{e\alpha}_{\vec{q}}$ on the conserved densities $X^3_{\vec{q}}$, $X^4_{\vec{q}}$ vanish.[10] Therefore, in the expansion (2.11) of the currents, only nonconserved operators contribute. This implies that the total current is nonconserved, or (so to speak) dissipative, and appears in the transport coefficients. Using the conservation laws of the normalized operators

$$\dot{X}^3_{\vec{q}} = -iq^{\alpha}\langle M_{\vec{q}}, M_{\vec{q}}\rangle^{-1/2} j^{M\alpha}_{\vec{q}} \ ,$$

$$\dot{X}^4_{\vec{q}} = -iq^{\alpha}[1/d(\vec{q})](\langle M_{\vec{q}}, M_{\vec{q}}\rangle j^{e\alpha}_{\vec{q}} \qquad (3.15)$$

$$- \langle \mathcal{H}^e_{\vec{q}}, M_{\vec{q}}\rangle j^{M\alpha}_{\vec{q}}) \ ,$$

the hydrodynamic equations may be inferred from Eqs. (2.11) and (2.15)–(2.18):

$$\delta\dot{X}^3_{\vec{q}} = -q^{\alpha}q^{\beta}D^{33}_{\alpha\beta}\,\delta X^3_{\vec{q}} - q^{\alpha}q^{\beta}D^{34}_{\alpha\beta}\,\delta X^4_{\vec{q}} \ ,$$

$$\delta\dot{X}^4_{\vec{q}} = -q^{\alpha}q^{\beta}D^{43}_{\alpha\beta}\,\delta X^3_{\vec{q}} - q^{\alpha}q^{\beta}D^{44}_{\alpha\beta}\,\delta X^4_{\vec{q}} \ . \qquad (3.16)$$

The transport coefficients, which are in general q dependent are given by the Kubo formulas:

1. Spin diffusion:

$$D^{33}(\vec{q}) = (1/\langle M_{\vec{q}}, M_{\vec{q}}\rangle$$

$$\times \operatorname{Lim} \tfrac{1}{2}\beta \int_{-\infty}^{\infty} dt \, \langle \tfrac{1}{2}\{j^{M\alpha}_{\vec{q}}(t), j^{M\beta}_{\vec{q}}(0)^{\dagger}\}\rangle_c e^{i\omega t} \ . \qquad (3.17)$$

More precisely, according to the first line of Eq. (2.15) we should apply Lim to the normalized currents $j^M_{\vec{q}}\langle j^M_{\vec{q}}, j^M_{\vec{q}}\rangle^{-1/2}$. In Eq. (3.17) this amounts to a factor $\langle j^M_{\vec{q}}, j^M_{\vec{q}}\rangle^{-1} \operatorname{Lim}\langle j^M_{\vec{q}}, j^M_{\vec{q}}\rangle$, which, as long as $\langle j^M_{\vec{q}}, j^M_{\vec{q}}\rangle$ is not singular for $q \to 0$, is set equal to 1 in the order in which we are interested. A similar remark applies also to the other diffusion constants. If one is interested in higher nonlocal terms in the hydrodynamic equations, the dependence on $\vec{q}$ of such factors has also to be taken into account. (See also Ref. 8.)

2. Thermal diffusion:

$$D^{44}_{\alpha\beta}(\vec{q}) = d(\vec{q})^{-2}[\langle M_{\vec{q}}, M_{\vec{q}}\rangle^2$$

$$\times \operatorname{Lim} \tfrac{1}{2}\beta \int_{-\infty}^{\infty} dt \, \langle \tfrac{1}{2}\{j^{e\alpha}_{\vec{q}}(t), j^{e\beta}_{\vec{q}}(0)^{\dagger}\}\rangle_c e^{i\omega t}$$

$$- 2\operatorname{Re}\langle M_{\vec{q}}, M_{\vec{q}}\rangle \langle \mathcal{H}^e_{\vec{q}}, M_{\vec{q}}\rangle$$

$$\times \operatorname{Lim} \tfrac{1}{2}\beta \int_{-\infty}^{\infty} dt \, \langle \tfrac{1}{2}\{j^{M\alpha}_{\vec{q}}(t), j^{e\beta}_{\vec{q}}(0)^{\dagger}\}\rangle_c e^{i\omega t}$$

$$+ |\langle \mathcal{H}^e_{\vec{q}}, M_{\vec{q}}\rangle|^2$$

$$\times \operatorname{Lim} \tfrac{1}{2}\beta \int_{-\infty}^{\infty} dt \, \langle \tfrac{1}{2}\{j^{M\alpha}_{\vec{q}}(t), j^{M\beta}_{\vec{q}}(0)^{\dagger}\}\rangle_c e^{i\omega t}] \ . \qquad (3.18)$$

3. Thermomagnetic diffusion:

$$D^{34}_{\alpha\beta}(\vec{q}) = d(\vec{q})^{-1}[(\langle M_{\vec{q}}, M_{\vec{q}}\rangle)^{1/2}$$

$$\times \operatorname{Lim} \tfrac{1}{2}\beta \int_{-\infty}^{\infty} dt\, e^{i\omega t} \langle \tfrac{1}{2}\{j^{M\alpha}_{\vec{q}}(t), j^{e\beta}_{\vec{q}}(0)^{\dagger}\}\rangle_c$$

$$- \langle \mathcal{H}^e_{\vec{q}}, M_{\vec{q}}\rangle / (\langle M_{\vec{q}}, M_{\vec{q}}\rangle)^{1/2}$$

$$\times \operatorname{Lim} \tfrac{1}{2}\beta \int_{-\infty}^{\infty} dt\, e^{i\omega t} \langle \tfrac{1}{2}\{j^{M\alpha}_{\vec{q}}(t), j^{M\beta}_{\vec{q}}(0)^{\dagger}\}\rangle_c] \ . \qquad (3.19)$$

In crystals with cubic symmetry or in isotropic systems these tensors are proportional to the unit matrix

$$D^{ij}_{\alpha\beta} = D^{ij}\,\delta_{\alpha\beta} \ . \qquad (3.20)$$

We also note that $D^{ij}_{\alpha\beta}$ has the properties of a scalar product. This implies, besides linearity, that $D^{ii}_{\alpha\alpha}$ is real and $\geqslant 0$, that $D^{ij*}_{\alpha\beta} = D^{ji}_{\beta\alpha}$, and that the Schwarz inequality,

$$|D^{ij}_{\alpha\beta}|^2 \leqslant D^{ii}_{\alpha\alpha} D^{jj}_{\beta\beta} \ , \qquad (3.21)$$

holds.

The coupled diffusion Eq. (3.16) which, by use of Eqs. (3.12) and (3.13), can be transformed into diffusion equations for the unnormalized densities, $M_{\vec{q}}$, $\mathcal{H}^e_{\vec{q}}$, have the following eigenfrequencies:

$$\omega'_{\pm} = -iD_{\pm}q^2$$

$$= -\tfrac{1}{2} i q^2 \{D^{33} + D^{44} \pm [(D^{33} - D^{44})^2 + 4|D^{34}|^2]^{1/2}\} . \qquad (3.22)$$

Here we have limited ourselves to a lattice of cubic symmetry [see Eq. (3.20)]. For stability the diffusion constants $D_{\pm}$ must be positive; this is guaranteed by the Schwarz inequality (3.21). The absorptive part of the dynamic susceptibility is given by

$$\chi''^{MM}_{(\vec{q},\omega)} = \chi^{MM}_{(\vec{q},\omega=0)}$$

$$\times \left\{ \frac{\omega D_+ q^2}{\omega^2 + (D_+ q^2)^2} \frac{D_+ - D^{44}}{D_+ - D_-} + \frac{\omega D_- q^2}{\omega^2 + (D_- q^2)^2} \frac{-D_- + D^{44}}{D_+ - D_-} \right\} . \qquad (3.23)$$

This can be related to the dynamic form factor by Eq. (6.10). Above the Curie point, if there is no external field, $\langle \mathcal{H}^e_{\vec{q}}, S^z_{\vec{q}} \rangle$ vanishes. In this case the hydrodynamic equations reduce to

$$\frac{d}{dt}\delta S^{\nu}_{\vec{q}} = -q^{\alpha} q^{\beta} D^{M}_{\alpha\beta} \delta S^{\nu}_{\vec{q}} \quad , \tag{3.24}$$

and
$$\frac{d}{dt}\delta \mathcal{H}^{e}_{\vec{q}} = -q^{\alpha} q^{\beta} D^{e}_{\alpha\beta}\, \delta \mathcal{H}^{e}_{\vec{q}} \quad . \tag{3.25}$$

The diffusion constants are found from Eqs. (3.17)–(3.19) by taking the limit

$$\langle \mathcal{H}^e_{\vec{q}}, S^z_{\vec{q}} \rangle \rightarrow 0 \ .$$

The cross term D^{34} also vanishes.

C. Hydrodynamics with Momentum Conservation

Every continuous symmetry implies a conservation law (Noether's theorem). Since the Hamiltonian (3.1) is translationally invariant only with respect to a lattice vector, the generator of this transformation, the (quasi-) momentum will not in general be conserved. It is physically clear and well known that momentum is approximately conserved at low temperatures.

In order to study effects connected with translation invariance it is convenient to introduce a boson representation by means of the Holstein-Primakoff transformation[11]

$$\begin{aligned} S^{+}_{\vec{l}} &= (2S)^{1/2}[1-(1/2S)a^{\dagger}_{\vec{l}} a_{\vec{l}}]^{1/2} a_{\vec{l}} \ , \\ S^{-}_{\vec{l}} &= (2S)^{1/2} a^{\dagger}_{\vec{l}} [1-(1/2S)a^{\dagger}_{\vec{l}} a_{\vec{l}}]^{1/2} \ , \\ S^{z}_{\vec{l}} &= S - a^{\dagger}_{\vec{l}} a_{\vec{l}} \ . \end{aligned} \tag{3.26}$$

The commutation relations for the boson operators $a_{\vec{l}}$ are

$$[a_{\vec{l}}, a^{\dagger}_{\vec{l}'}] = \delta_{\vec{l},\vec{l}'} \ , \quad [a_{\vec{l}}, a_{\vec{l}'}] = 0 \ . \tag{3.27}$$

Let us introduce the Fourier-transformed variables

$$a^{\dagger}_{\vec{k}} = N^{-1/2} \textstyle\sum_{\vec{l}} e^{i\vec{k}\cdot\vec{x}_{\vec{l}}}\, a^{\dagger}_{\vec{l}} , \quad a_{\vec{k}} = N^{-1/2} \textstyle\sum_{\vec{l}} e^{-i\vec{k}\cdot\vec{x}_{\vec{l}}}\, a_{\vec{l}} ,$$
$$\vec{k} = \sum_{\alpha=1}^{3} \vec{b}_{\alpha} \frac{\nu_{\alpha}}{N_{\alpha}} \ . \tag{3.28}$$

We imagine we have a cubic crystal with conventional lattice constant a. The number of atoms in the direction of the basis vector $\vec{a}_{\alpha}$ is N_{α}, and the total number of atoms $N = N_1 N_2 N_3$. The reciprocal lattice is spanned by $\vec{b}_{\beta}$ ($\vec{b}_{\beta}\cdot\vec{a}_{\alpha} = 2\pi\,\delta_{\beta\alpha}$). The integer ν_{α} varies between $-\frac{1}{2}N_{\alpha}$ and $\frac{1}{2}N_{\alpha}$. The operators $a_{\vec{k}}$ obey the commutation relations

$$[a_{\vec{k}}, a^{\dagger}_{\vec{k}'}] = \Delta(\vec{k}-\vec{k}') \ . \tag{3.29}$$

The momentum density may be written

$$P^{\alpha}_{\vec{q}} = \textstyle\sum_{\vec{k}} k^{\alpha}\, a^{\dagger}_{\vec{k}-(\vec{q}/2)} a_{\vec{k}+(\vec{q}/2)} \ . \tag{3.30}$$

The generalized Kronecker $\delta\,\Delta(\vec{p})$ is defined to vanish except when its argument is zero or a reciprocal-lattice vector, in which case it equals unity.

The advantage of having bosons without kinematical interaction is paid for by the presence of the square root in (3.26), which gives rise to nonlinearities of infinite order in the Hamiltonian. Oguchi[12] has established that one gets Dyson's[4] result up to order $(1/S)^0$ if one keeps all terms up to order $(1/S)^0$.[13] Similarly, we make here an expansion in $1/S$ keeping only terms up to order $(1/S)^0$. The structure of the hydrodynamic equations does not depend on where one truncates the expansion in $1/S$ of the Hamiltonian. Only if one wants to calculate explicitly the coefficients in the hydrodynamic equations, does this come into play. One also can derive the hydrodynamic equations for the case of spin $\frac{1}{2}$ without any approximation. This will be published in a forthcoming communication.

Through order $(1/S)^0$ the Hamiltonian reads

$$\begin{aligned} \mathcal{H} = &\sum_{\vec{k}} (\omega^0_{\vec{k}} + g\mu_B H) a^{\dagger}_{\vec{k}} a_{\vec{k}} \\ &+ \frac{1}{2N} \sum_{\vec{k}_1,\ldots\vec{k}_4} W(\vec{k}_1,\vec{k}_2,\vec{k}_3,\vec{k}_4)\, \Delta(\vec{k}_1+\vec{k}_2-\vec{k}_3-\vec{k}_4) \\ &\times a^{\dagger}_{\vec{k}_1} a^{\dagger}_{\vec{k}_2} a_{\vec{k}_3} a_{\vec{k}_4} - g\mu_B HNS \ . \end{aligned} \tag{3.31}$$

In (3.31), the harmonic-magnon energy is

$$\omega^0_{\vec{k}} = S(\tilde{J}(0) - \tilde{J}(\vec{k})) \ , \tag{3.32}$$

$$\tilde{J}(\vec{k}) = \textstyle\sum_{\vec{l}} e^{i\vec{k}\cdot\vec{x}_{\vec{l}}}\, J(\vec{l}) \tag{3.33}$$

and the four-magnon interaction potential reads

$$W(\vec{k}_1,\vec{k}_2,\vec{k}_3,\vec{k}_4) = \tfrac{1}{2}\tilde{J}(\vec{k}_1) + \tfrac{1}{2}\tilde{J}(\vec{k}_4) - \tilde{J}(\vec{k}_1-\vec{k}_4) \ . \tag{3.34}$$

In addition to the conservation laws (3.8) and (3.9) for magnetization and energy, we have in the present low-temperature case

$$\dot{P}^{\alpha}_{\vec{q}} = -iq^{\beta}\,\Pi^{\alpha\beta}_{\vec{q}} - i\,u^{\alpha}_{\vec{q}} \ . \tag{3.35}$$

The momentum flux operator is given by

$$\begin{aligned} \Pi^{\alpha\beta}_{\vec{q}} = &\sum_{\vec{k}} k^{\alpha} \frac{\partial \omega^0_k}{\partial k^{\beta}} a^{\dagger}_{\vec{k}-(\vec{q}/2)} a_{\vec{k}+(\vec{q}/2)} \\ &+ \frac{1}{4N} \sum_{\vec{k}_1,\ldots\vec{k}_4} a^{\dagger}_{\vec{k}_1} a^{\dagger}_{\vec{k}_2} a_{\vec{k}_3} a_{\vec{k}_4}\, \Delta(\vec{q}+\vec{k}_1+\vec{k}_2-\vec{k}_3-\vec{k}_4) \\ &\times \Bigg[\frac{\partial}{\partial k_1^{\beta}} k_1^{\alpha} \tilde{J}(\vec{k}_1) + \frac{\partial}{\partial k_4^{\beta}} k_4^{\alpha} \tilde{J}(\vec{k}_4) \\ &- 2\frac{\partial}{\partial k_1^{\beta}} (k_1^{\alpha} - k_4^{\alpha}) \tilde{J}(\vec{k}_1-\vec{k}_4) \Bigg] \ . \end{aligned} \tag{3.36}$$

Because of the discreteness of the lattice, there is a term in (3.35) which is not proportional to q:

$$\begin{aligned} u^{\alpha}_{\vec{q}} = \tfrac{1}{4} &\sum_{\vec{k}_1,\ldots\vec{k}_4} \Delta(\vec{q}+\vec{k}_1+\vec{k}_2-\vec{k}_3-\vec{k}_4) g^{\alpha} \\ &\times (\vec{q}+\vec{k}_1+\vec{k}_2-\vec{k}_3-\vec{k}_4)[\tilde{J}(\vec{k}_1)+\tilde{J}(\vec{k}_4)-2\tilde{J}(\vec{k}_1-\vec{k}_4)] \\ &\times a^{\dagger}_{\vec{k}_1} a^{\dagger}_{\vec{k}_2} a_{\vec{k}_3} a_{\vec{k}_4} \ . \end{aligned} \tag{3.37}$$

The symbol $g^\alpha(\vec{k})$ denotes a reciprocal-lattice vector which is defined such that in the relation

$$k^\alpha = k_B^\alpha + g^\alpha(\vec{k}) \quad , \tag{3.38}$$

the vector k_B^α lies in the first Brillouin zone. In the derivation of (3.35)–(3.38), we used

$$[P_{\vec{q}}^\alpha, a_{\vec{k}}] = -[k^\alpha + \tfrac{1}{2}q^\alpha - g^\alpha(\vec{k} + \tfrac{1}{2}\vec{q})]\,a_{\vec{k}+\vec{q}} \quad . \tag{3.39}$$

The vector $u_{\vec{q}}^\alpha$ is different from zero only for umklapp processes. Therefore its scalar product with any other operator is of order $\kappa = a^{-1}e^{-T_D/T}$, where T_D is the Debye temperature of the magnons. There is yet another contribution to (3.35) arising from the boundary of the Brillouin zone and containing a factor of the type $g^\alpha(\vec{k}_s + \vec{q})$. Since it is proportional to q and different from zero only for $\vec{k}_s$ at the zone boundary, it is of the order $\vec{q}\cdot\kappa$ and therefore negligible compared to the linear terms.

The expectation value of the momentum flux operators (3.36) defines a pressure P,

$$PV\,\delta^{\alpha\beta} = \langle \Pi_{\vec{q}=0}^{\alpha\beta} \rangle \quad . \tag{3.40}$$

In fact, this pressure has also thermodynamic significance. In the low-temperature limit where magnons at the boundary of the Brillouin zone are not excited, P is the volume derivative of the internal energy at constant entropy and constant $M - M_0$. Since we do not need this property in the derivation of the hydrodynamic equations we have deferred the proof to Appendix A.

For the normalization of the momentum operator $P_{\vec{q}}^\alpha$ we need $\langle P_{\vec{q}}^\alpha, P_{\vec{q}}^\beta \rangle$. In an isotropic or cubic system, this will be

$$\langle P_{\vec{q}}^\alpha, P_{\vec{q}}^\beta \rangle = V\rho\,\delta^{\alpha\beta} + c q^\alpha q^\beta + O(q^4) \quad , \tag{3.41}$$

where ρ and c are constants. (ρ has nothing to do with the material density of the system.) If we neglect terms of $O(q^2)$ the normalized momentum, is

$$X_{\vec{q}}^{5\alpha} = (V\rho)^{-1/2} P_{\vec{q}}^\alpha \quad . \tag{3.42}$$

In a spatially more anisotropic crystal, the different components of the momentum will no longer be orthogonal. It is not hard to extend the derivation to such systems.

After these preliminaries let us now deduce the hydrodynamics from the conservation laws (3.8), (3.9), and (3.37). The currents $j_{\vec{q}}^{M\alpha}$ and $j_{\vec{q}}^{e\alpha}$ have finite projections on the momentum operator. The projection of $\Pi_{\vec{q}}^{\alpha\beta}$ on $\mathcal{H}_{\vec{q}}^e$ and $M_{\vec{q}}$ is also nonzero. The scalar products of the currents in (3.8), (3.9), and (3.35) with the transverse spin components are still zero. For the calculation of the projections, it is useful to employ the identity

$$\langle \dot{A}_{\vec{q}}, B_{\vec{q}} \rangle = -i\langle [A_{\vec{q}}, B_{\vec{q}}^\dagger] \rangle \quad , \tag{3.43}$$

which follows from (2.5) by partial integration. So we find

$$\langle j_{\vec{q}}^{M\alpha}, P_{\vec{q}}^\beta \rangle = \delta^{\alpha\beta}(M - M_0) \quad , \tag{3.44}$$

$$\langle j_{\vec{q}}^{e\alpha}, P_{\vec{q}}^\beta \rangle = \delta^{\alpha\beta}(U + PV) \quad . \tag{3.45}$$

In (3.44) and (3.45), M is the equilibrium magnetization, M_0 the saturation magnetization, and U the internal energy (see Appendix A), and PV is given by (3.40). From Eqs. (3.44) and (3.45) follows the decomposition of the currents in a conserved part and in a nonconserved part $\tilde{j}$:

$$j_{\vec{q}}^{M\alpha} = [(M - M_0)/(V\rho)^{1/2}]\,X_{\vec{q}}^{5\alpha} + \tilde{j}_{\vec{q}}^{M\alpha} \quad , \tag{3.46}$$

$$j_{\vec{q}}^{e\alpha} = [(U + PV)/(V\rho)^{1/2}]\,X_{\vec{q}}^{5\alpha} + \tilde{j}_{\vec{q}}^{e\alpha} \quad . \tag{3.47}$$

The decomposition of the momentum flux operator $\Pi_{\vec{q}}^{\alpha\beta}$ will be found by use of the symmetry condition (2.10). The conservation laws for the normalized operators are

$$\begin{aligned}
\dot{X}_{\vec{q}}^3 &= iq^\alpha c_3(\vec{q})X_{\vec{q}}^{5\alpha} - iq^\alpha \langle M_{\vec{q}}, M_{\vec{q}} \rangle^{-1/2}\tilde{j}_{\vec{q}}^{M\alpha} \quad , \\
\dot{X}_{\vec{q}}^4 &= -iq^\alpha c_4(\vec{q})X_{\vec{q}}^{5\alpha} - iq^\alpha d(\vec{q})^{-1} \\
&\quad \times (\langle M_{\vec{q}}, M_{\vec{q}} \rangle \tilde{j}_{\vec{q}}^{e\alpha} - \langle \mathcal{H}_{\vec{q}}^e, M_{\vec{q}} \rangle \tilde{j}_{\vec{q}}^{M\alpha}) \quad , \\
\dot{X}_{\vec{q}}^{5\alpha} &= iq^\alpha c_3(\vec{q})X_{\vec{q}}^3 - iq^\alpha c_4(\vec{q})X_{\vec{q}}^4 \\
&\quad - iq^\beta (V\rho)^{-1/2}\,\tilde{\Pi}_{\vec{q}}^{\alpha\beta} - i(V\rho)^{-1/2}u_{\vec{q}}^\alpha \quad .
\end{aligned} \tag{3.48}$$

The dissipative part of $\Pi_{\vec{q}}$ is given by

$$\tilde{\Pi}_{\vec{q}}^{\alpha\beta} = \Pi_{\vec{q}}^{\alpha\beta} - \delta^{\alpha\beta}(V\rho)^{1/2}(-c_3(\vec{q})X_{\vec{q}}^3 + c_4(\vec{q})X_{\vec{q}}^4) \quad , \tag{3.49}$$

and the velocities $c_3(\vec{q})$ and $c_4(\vec{q})$ are

$$c_3(\vec{q}) = (M_0 - M)(\langle M_{\vec{q}}, M_{\vec{q}} \rangle V\rho)^{-1/2} \tag{3.50}$$

and

$$\begin{aligned}
c_4(\vec{q}) &= [1/(V\rho)^{1/2}d(\vec{q})]\,(\langle M_{\vec{q}}, M_{\vec{q}} \rangle (U + PV) \\
&\quad - \langle \mathcal{H}_{\vec{q}}^e, M_{\vec{q}} \rangle (M - M_0)) \quad .
\end{aligned} \tag{3.51}$$

From Eqs. (3.48) and (2.15)–(2.19) we find immediately the hydrodynamic equations:

$$\delta\dot{X}_{\vec{q}}^3 = iq^\alpha c_3(q)\,\delta X_{\vec{q}}^{5\alpha} - q^2\tilde{D}^{33}(q)\,\delta X_{\vec{q}}^3 - q^2\tilde{D}^{34}(q)\delta X_{\vec{q}}^4 \quad , \tag{3.52a}$$

$$\begin{aligned}
\delta\dot{X}_{\vec{q}}^4 &= -iq^\alpha c_4(q)\delta X_{\vec{q}}^{5\alpha} \\
&\quad - q^2\tilde{D}^{43}(q)\delta X_{\vec{q}}^3 - q^2\tilde{D}^{44}(q)\delta X_{\vec{q}}^4 \quad ,
\end{aligned} \tag{3.52b}$$

$$\begin{aligned}
\delta\dot{X}_{\vec{q}}^{5\alpha} &= -iq^\alpha(-c_3(q)\delta X_{\vec{q}}^3 + c_4(q)\delta X_{\vec{q}}^4) \\
&\quad - q^\sigma q^\tau \tilde{D}_{\alpha\sigma\beta\tau}^{5\ 5}\,\delta X_{\vec{q}}^{5\beta} - \omega_P\,\delta X_{\vec{q}}^{5\alpha} \quad .
\end{aligned} \tag{3.52c}$$

We have indicated that the velocities and diffusion "constants" themselves will in general be q dependent. The hydrodynamic Eqs. (3.52) are identical to the ones obtained from the Boltzmann equation[14] [Eq. (3.14)]; one has to identify $\delta X_{\vec{q}}^3 \equiv \mathcal{M}(\vec{q}, t)$, $\delta X_{\vec{q}}^4 \equiv \boldsymbol{\vartheta}(\vec{q}, t)$, and $\delta X_{\vec{q}}^5 \equiv \mathcal{P}(\vec{q}, t)$. If one uses the definitions (3.6), (3.7), and (3.42) one may rewrite Eqs. (3.52) into a set of equations for the unnormalized densities (magnetization, momentum, and energy or temperature).

The hydrodynamic equations for the ferromagnet (3.52) contain a variety of transport coefficients. The diffusion constants $\tilde{D}^{33}$, $\tilde{D}^{34}$, and $\tilde{D}^{44}$ have already been defined in Eqs. (3.17)–(3.19). The only difference is that we have to replace $j_{\vec{q}}^{c\alpha}$ by $\tilde{j}_{\vec{q}}^{c\alpha}$. We note that $\tilde{D}^{44}$ is the nonconvective part of the thermal diffusivity. Because part of the energy current is proportional to the momentum density, the bulk magnon thermal conductivity diverges if we leave aside umklapp processes and impurities. In a liquid experimentally one can avoid convection and measure $\tilde{D}^{44}$; in a magnet a convective flux is built up in such an experiment, if there are number-violating interactions on the surface. The viscosity is given by

$$\tilde{D}^{5\;5}_{\alpha\sigma\beta\tau} = (1/V\rho)\,\mathrm{Lim}\,\tfrac{1}{2}\beta\int_{-\infty}^{\infty} dt\,\langle\tfrac{1}{2}\{\tilde{\Pi}_{\vec{q}}^{\alpha\sigma}(t), \tilde{\Pi}_{\vec{q}}^{\beta\tau\dagger}(0)\}\rangle_c\, e^{i\omega t}. \tag{3.53}$$

In a cubic crystal $\tilde{D}^{55}$ decomposes into

$$\tilde{D}^{5\;5}_{\alpha\sigma\beta\tau} = (\zeta - \tfrac{2}{3}\eta)\delta^{\alpha\sigma}\delta^{\beta\tau} + \eta(\delta^{\alpha\beta}\delta^{\sigma\tau} + \delta^{\alpha\tau}\delta^{\beta\sigma}) + \zeta'\delta^{\alpha\sigma\beta\tau} \quad . \tag{3.54}$$

The tensor $\delta_{\alpha\beta\sigma\tau}$ is different from zero only for $\alpha=\beta=\sigma=\tau$. In an isotropic medium the coefficient ζ' vanishes; the viscosity η is given by the quadrupole contribution to $\Pi_{\vec{q}}^{\alpha\beta}$ and the bulk viscosity ζ by the scalar part. The presence of the momentum relaxation rate due to umklapp processes ω_P in (3.48) is typical for a discrete system

$$\omega_P\delta^{\alpha\beta} = \frac{1}{V\rho}\lim_{\omega\to 0}\lim_{q\to 0}\frac{\beta}{2}\int_{-\infty}^{\infty} dt \times e^{i\omega t}\langle\tfrac{1}{2}\{u_{\vec{q}}^{\alpha}(t), u_{\vec{q}}^{\beta\dagger}(0)\}\rangle_c \quad . \tag{3.55}$$

Off-diagonal diffusion constants like $\tilde{D}^{35}$, $\tilde{D}^{45}$ are zero in isotropic or cubic crystals. For noncubic anisotropic lattices the momentum susceptibility $\langle P_{\vec{q}}^{\alpha}, P_{\vec{q}}^{\beta}\rangle$ is a tensor. The transformation matrix which diagonalizes this tensor appears in the velocities and Kubo formulas.

There are still other dissipative terms like $-q^{\alpha}\mu^{35}_{\alpha\beta}\delta X_{\vec{q}}^{5\beta}$ in $\delta\dot{X}_{\vec{q}}^{3}$ and similar terms in $\delta\dot{X}_{\vec{q}}^{6}$ and $\delta\dot{X}_{\vec{q}}^{5\alpha}$. The coefficient $q^{\alpha}\mu^{35}_{\alpha\beta}$ is proportional to

$$q^{\alpha}\int_{-\infty}^{\infty} dt\,\langle\tfrac{1}{2}\{\tilde{j}_{\vec{q}}^{M\alpha}(t),\ u_{\vec{q}}^{\beta\dagger}(0)\}\rangle_c\,, \tag{3.56}$$

and is estimated to be of order $cq\omega_P/\omega_N$ and therefore neglected compared to the diffusion terms.

It can be seen from the hydrodynamic Eqs. (3.52) that the system may perform a wavelike motion with velocity $[c_3^2(q)+c_4^2(q)]^{1/2}$ if the damping terms are small.

One would also expect that (3.52) reduces to the hydrodynamic Eqs. (3.16) if ω_P becomes large, i.e., $\omega_P \approx \omega_N$. In fact, for large ω_P, (3.52c) can be written as

$$-iq^{\alpha}[-c_3(q)\,\delta X_{\vec{q}}^{3} + c_4(q)\,\delta X_{\vec{q}}^{4}] - \omega_P\,\delta X_{\vec{q}}^{5\alpha} = 0. \tag{3.57}$$

Inserting (3.57) into (3.52a) and (3.52b) gives

$$\begin{aligned}\delta\dot{X}_{\vec{q}}^{3} &= -q^2[\tilde{D}^{33} + (c_3^2/\omega_P)]\,\delta X_{\vec{q}}^{3} - q^2[\tilde{D}^{34} - (c_3c_4/\omega_P)]\,\delta X_{\vec{q}}^{4},\\ \delta\dot{X}_{\vec{q}}^{4} &= -q^2[\tilde{D}^{43} - (c_3c_4/\omega_P)]\,\delta X_{\vec{q}}^{3} - q^2[\tilde{D}^{44} + (c_4^2/\omega_P)]\,\delta X_{\vec{q}}^{4}.\end{aligned} \tag{3.58}$$

The additional diffusive terms make up for the difference in definition of D^{ij} and $\tilde{D}^{ij}$. Equation (3.58) applies also if $\omega \ll \omega_P$ and arbitrary ω_p.

Finally we remark that the structure of the hydrodynamic equations is unchanged if the exchange interaction is anisotropic, as long as $\sum S_{\vec{l}}^{z}$ is conserved. Only the values of the coefficients will depend on these details. In Appendix B we give the modifications resulting from anisotropic exchange interaction.

IV. SOLUTIONS OF HYDRODYNAMIC EQUATIONS ($H \neq 0$); MODES AND RESPONSE FUNCTIONS

In the following we study the characteristic modes of the hydrodynamic equations (3.52). We recall that $\delta X_{\vec{q}}^{3}$, $\delta X_{\vec{q}}^{4}$, and $\delta X_{\vec{q}}^{5\alpha}$ are, except for a normalization factor, equal, respectively, to the local magnetization, temperature, and momentum [Eqs. (3.12), (3.13), and (3.42)]. Let us for the sake of definiteness imagine that the wave vector q lies in the [1, 0, 0] direction. Use of expression (3.54) for the viscosity tensor in (3.52c) shows that the transverse components of the normalized momentum, $\delta X_{\vec{q}}^{52}$ and $\delta X_{\vec{q}}^{53}$, are uncoupled to the rest of the system. The corresponding eigensolution is the viscous mode

$$\delta X_{\vec{q}}^{5\alpha}(t) = \delta X_{\vec{q}}^{5\alpha}(0)\,e^{-\Gamma_v t}, \quad \alpha = 2, 3, \qquad \Gamma_v = \eta q^2 + \omega_P\,. \tag{4.1}$$

The remaining set of hydrodynamic equations is made up of the z component of magnetization, temperature, and longitudinal momentum, and may be written

$$[\omega 1 - q^{\alpha}m_{\alpha}(\vec{q}) + i\Gamma(\vec{q})]^{cc'}\begin{Bmatrix}\delta X_{\vec{q}}^{c'}\\ \Phi^{c'c''}(\vec{q},\omega)\end{Bmatrix} = \begin{Bmatrix}0\\ \delta^{cc''}\end{Bmatrix}, \tag{4.2a, 4.2b}$$

where the matrix $-q^{\alpha}m_{\alpha} + i\Gamma$ is given by

$$-q^{\alpha}m_{\alpha} + i\Gamma = \begin{pmatrix} i\tilde{D}^{33}q^2 & iD^{34}q^2 & c_3q\\ i\tilde{D}^{34*}q^2 & i\tilde{D}^{44}q^2 & -c_4q\\ c_3q & -c_4q & i(\tfrac{4}{3}\eta + \zeta + \zeta')q^2 + i\omega_P\end{pmatrix}. \tag{4.3}$$

The eigenfrequencies are found from

$$\mathrm{Det}(\omega 1 - q^{\alpha}m_{\alpha} + i\Gamma) = 0 \quad , \tag{4.4}$$

and the relaxation functions by inverting (4.2b), e.g., the magnetization-magnetization relaxation function is given by

$$\Phi^{zz}(q,\omega) \equiv \Phi^{MM}(q,\omega) = \chi^{zz}(q,0)$$

$$\times \frac{i\{(\omega + iD_{44}q^2)[\omega + i(\frac{4}{3}\eta + \zeta + \zeta')q^2 + i\omega_P] - c_4^2 q^2\}}{\operatorname{Det}(\omega 1 - q^\alpha m_\alpha + i\Gamma)} \quad . \tag{4.5}$$

Usually one takes the limit $q \to 0$ of the susceptibilities entering in the coefficients of the hydrodynamic equations. Since the longitudinal susceptibility for the isotropic Heisenberg ferromagnet is divergent for q and $H \to 0$, the limit may be performed only in the presence of an external or anisotropy field and only for $q \ll q_c$, where the characteristic wave number q_c is given by

$$q_c = a^{-1}(g\mu_B H/JS)^{1/2} \quad , \tag{4.6}$$

as will become clear in Sec. V. Now we suppose that we have an external field or anisotropy such that $q \ll q_c$, as will be the case in most practical situations, and take the thermodynamic limit. In the derivation of the hydrodynamic equations one makes an expansion in q, which implies the condition $c_{3,4}q > D^{ij}q^2$. Employing a collision time approximation for D^{ij} this condition may be rewritten in the form

$$cq < c^2/D^{ij} \approx \omega_N \quad . \tag{4.7}$$

The nature of the modes derived from (4.3) depends crucially on the relative magnitude of the momentum-relaxation frequency ω_P and $c_{3,4}q$. We may distinguish four different regimes.

(i) $\omega_P \ll c_s q \ll \omega_N$: Frequency window[15] and $c_s\, q$ within the window.

One finds from (4.4) a diffusive mode

$$\omega_1 = -iD_T q^2$$

$$\equiv (-i/c_s^2)(2\operatorname{Re}\tilde{D}^{34}c_3c_4 + \tilde{D}^{33}c_4^2 + \tilde{D}^{44}c_3^2)q^2 \quad , \tag{4.8}$$

and a damped propagating mode (*second magnon*)

$$\omega_\pm = \pm c_s q - \tfrac{1}{2} i (D_s q^2 + \omega_P) \quad , \tag{4.9}$$

where

$$c_s = (c_3^2 + c_4^2)^{1/2} \quad , \tag{4.10}$$

$$D_s = \tilde{D}^{33}(c_3^2/c_s^2) + \tilde{D}^{44}(c_4^2/c_s^2) + \tfrac{4}{3}\eta + \zeta + \zeta' - 2\operatorname{Re}\tilde{D}^{34}(c_3c_4/c_s^2) \quad . \tag{4.11}$$

The velocities c_3 and c_4 of Eqs. (3.50) and (3.51) may be written in terms of thermodynamic derivatives by means of (A8)–(A10):

$$c_3 = \left[\left(\frac{\partial M}{\partial H}\right)_{T,V} V\rho\right]^{-1/2}(M_0 - M) \quad ,$$

$$c_4 = (V\rho\, TC_{M,V})^{-1/2}$$

$$\times \left\{U + PV + \left[1 - T\left(\frac{\partial H}{\partial T}\right)_{M,V}\right](M_0 - M)\right\} . \tag{4.12}$$

Of particular interest is the oscillatory solution, the second magnon. In order to determine whether it is primarily a temperature wave or an oscillation of the z component of the magnetization, we have to find the amplitude of $M_{\vec{q}}$ in this mode or equivalently the residue of the pole (4.9) in the magnetization correlation function. From Eqs. (4.5) and (2.17), we find for the longitudinal dynamic form factor [defined in Eq. (6.10)]

$$S^{zz}(\vec{q},\omega) = T\left(\frac{\partial M}{\partial H}\right)_T \left[\frac{q^2 D_T(1 - c_3^2/c_s^2)}{\omega^2 + (D_T q^2)^2} + \frac{q^2c_3^2(q^2D_s + \omega_P) - q^2[-\tilde{D}^{33} + D_T(1 - c_3^2/c_s^2)](\omega^2 - c_s^2q^2)}{(\omega^2 - c_s^2q^2)^2 + [\omega(D_s q^2 + \omega_P)]^2}\right] . \tag{4.13}$$

It is seen that the integrated strength of the diffusion pole in $S^{zz}(\vec{q},\omega)$ is $\frac{1}{2}T\,(\partial M/\partial H)_T(1 - c_3^2/c_s^2)$ and the integrated strength of each one of the propagating modes is $\frac{1}{4}(c_3^2/c_s^2)T(\partial M/\partial H)_T$. This is rather noteworthy because it shows that if $c_3^2 \approx c_s^2$, the propagating mode is mainly an oscillation of the z component of the magnetization. On the other hand, second sound in a phonon system and in liquid helium is mainly a temperature wave[16] coupled to the density fluctuation by the small ratio $(1 - c_T^2/c_s^2) = (c_P - c_V)/c_P$. The situation in the magnetic case is more analogous to a real gas where the propagating mode is mainly a density wave.[16] (The density corresponds to the z component of the magnetization.) The analogy can be pushed further if one applies the thermodynamic results discussed in the second half of Appendix A.

We find in (A20)–(A22), that

$$c_3^2 = -\frac{V}{\rho}\left(\frac{\partial P}{\partial V}\right)_{T,M-M_0}, \quad c_4^2 = \left[VT\left(\frac{\partial P}{\partial T}\right)^2_{M-M_0,V}\right] \Big/ \rho C_{M-M_0 V},$$

$$c_s^2 = -\frac{V}{\rho}\left(\frac{\partial P}{\partial V}\right)_{S,M-M_0} . \tag{4.14}$$

Here terms of order $e^{-T_D/T}$ have been neglected. The coupling of the propagating mode to the magnetization is $\propto (1 - c_{V,M-M_0}/c_{P,M-M_0})$. Its velocity is proportional to the adiabatic "magnetic compressibility." All these constants depend on the external field, by means of which one may vary the relative strength of propagating to diffusive mode and the velocities. In Appendix C we give values for these constants in the lowest-order magnon model. There we find that c_3^2/c_s^2 increases from 0 to $\frac{3}{5}$ if H varies from 0 to ∞. The velocities of the lowest-order magnon model agree with the results of the work by Reiter (Ref. 3), in which also an expression for the response function has been derived. In the damping of the second magnon we have included also the contribution due to thermal diffusion and umklapp processes.

If the wave number is lowered, the ratio of damping versus real part in (4.9) increases because of ω_P and we reach region (ii).

(ii) $\omega_P \approx c_s q \ll \omega_N$. The leading term of the "propagating" solutions is given by

$$\omega_\pm = -\tfrac{1}{2} i \omega_P \pm \left(-\tfrac{1}{4}\omega_P^2 + c_s^2 q^2\right)^{1/2} + O(q^2) \ . \qquad (4.15)$$

For $2c_s q = \omega_P$ the modes get critically damped, and for still smaller values of q we have only decaying modes. Which brings us into region (iii).

(iii) $c_s q \ll \omega_P \ll \omega_N$. In this region expression (4.15) reduces to $\omega_- \approx -i\omega_P$, $\omega_+ = -ic_s^2 q^2/\omega_P$. The correlation functions can be found from Eq. (4.5) in regions (ii) and (iii). If finally the temperature is so high that

(iv) $c_s q \ll \omega_P \approx \omega_N$, we have no reason to treat the momentum differently from any other fast-relaxing nonconserved operator. The coupled diffusion Eqs. (3.16) apply. (See also end of Sec. III.) The solutions in this region are purely diffusive and have been found already in Sec. III [Eq. (3.22)]. The absorptive part of the dynamical susceptibility has been given in Eq. (3.23). The over-all picture is the following:

Imagine we keep the wave number q fixed such that $c_s q \ll \omega_N$ and raise the temperature. Since ω_P increases, from zero to $\sim \omega_N$, we pass successively from regions (i)–(iv). We have illustrated this in Fig. 1, where we have shown the position of the poles in the complex ω plane. The arrows point in the direction of increasing temperature. At low temperatures, there is a propagating mode with damping $D_s q^2 + \omega_P$. With increasing temperature the poles move to the imaginary axis. One pole moves downward and disappears in the continuum of fast decaying oscillations. The other moves upward and becomes the diffusion pole $D_+ q^2$. In addition to the poles shown in the graph, there is always a diffusive peak present.

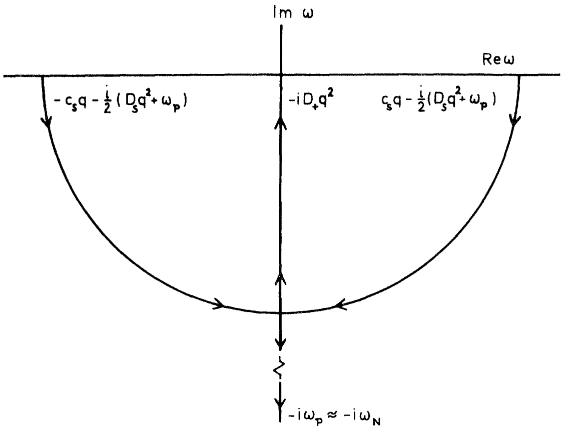

FIG. 1. Poles in the complex ω plane.

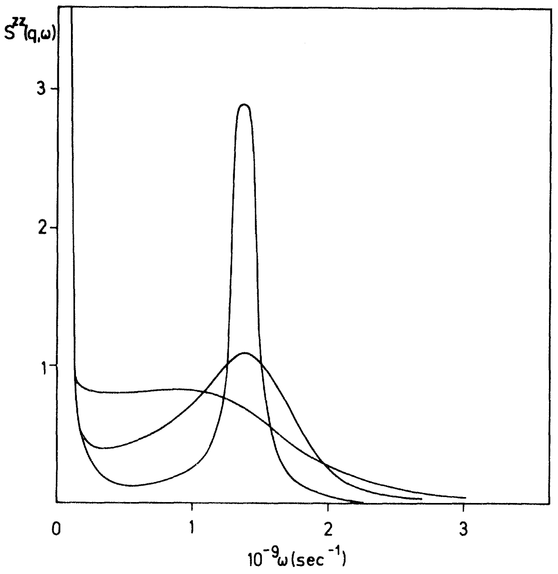

FIG. 2. Dynamic form factor $q = 10^{-4}$ Å^{-1}.

Figure 2 shows the dynamic form factor $S^{zz}(\vec{q}, \omega)$ for temperatures $4JS$, $5JS$, and $5.5JS$ and $H = 4JS/g\mu_B$ for the values in Appendix C. Since ω_P increases with temperature the propagating peak broadens. The integrated strength increases because the static susceptibility rises with T.

V. ISOTROPIC FERROMAGNET $H \to 0$ ($T \ll T_c$)

All coefficients in the hydrodynamic equations contain q-dependent susceptibilities. If the q dependence is weak and the limit $q \to 0$ is finite, this limit may be taken in the hydrodynamic regime. For instance, the energy susceptibility is given by

$$\langle \mathcal{H}^e_{\vec{q}}, \mathcal{H}^e_{\vec{q}} \rangle = \langle \mathcal{H}^e_0, \mathcal{H}^e_0 \rangle + O(q^2) \ .$$

Retaining the q dependence would require also that the Kubo formulas for the transport coefficients have to be generalized by iterating Eq. (2.12c').

In the following we shall treat wave numbers sufficiently small that one may perform $q \to 0$. However, there is one exception – the longitudinal magnetic susceptibility of an isotropic Heisenberg ferromagnet diverges at $q = 0$, $H = 0$. Therefore, the q dependence has to be retained here. In this section we shall first review the status of the longitudinal susceptibility and then discuss the consequences on the solutions of the hydrodynamic equations. In Sec. V C the transverse modes are treated.

A. Longitudinal Magnetic Susceptibility

It follows rigorously from Dyson's[4] theory that the derivative of M with respect to the external field H develops a $H^{-1/2}$ singularity for $H \to 0$. The leading term in an expansion in T is

$$\left(\frac{\partial M}{\partial H}\right)_T = (g\mu_B)^2 \frac{V T^{1/2}}{8\pi^{3/2}(JSa^2)^{3/2}} F_{1/2}\left(\frac{g\mu_B H}{T}\right) + O(T^{3/2}, T^3 F_{1/2}(g\mu_B H/T)) \quad , \tag{5.1a}$$

$$\left(\frac{\partial M}{\partial H}\right)_T = (g\mu_B)^2 \frac{V T^{1/2}}{8\pi(JSa^2)^{3/2}(g\mu_B H/T)^{1/2}} + O(H^0, H^{-1/2}T^{7/2}) \; . \tag{5.1b}$$

For notation see Appendix C. One expects that the static longitudinal susceptibility χ_q^{zz} for small values of q will be given roughly by replacing $g\mu_B H$ by JSq^2a^2. In fact, Mori and Kawasaki[17] have found a $1/q$ singularity for $JSa^2q^2 \gg g\mu_B H$ in the longitudinal susceptibility. Their result and the leading term (5.1b) are reproduced by the expression[18]

$$\chi_q^{zz} = (g\mu_B)^2 \frac{V T^{1/2}\psi}{8(JSa^2)^{3/2}\,(JSa^2q^2/T)^{1/2}} \quad , \tag{5.2}$$

where the function ψ is defined by

$$\psi = (2/\pi)\arctan(JSa^2q^2/4g\mu_B H)^{1/2}. \tag{5.3}$$

Eq. (5.2) has been arrived at by perturbation theory. Therefore the consequences which we shall draw from it are not as well founded as Sec. IV. It must be noted that, in addition to the singular terms (5.1) and (5.2), there are also nonsingular contributions to the susceptibility. We assume that the wave number and external field are so small that the singular terms dominate. At low temperatures, this will be the case for $qa < (T/JS)^{1/2}10^{-2}$ and $g\mu_B H/T < 10^{-2}$. In this range, χ_q^{zz} is much larger than the other susceptibilities; therefore we expand all coefficients in terms of $(g\mu_B)^2/\chi_q^{zz}$.

B. Modes and Response Functions

The velocities $c_3(q)$ and $c_4(q)$ are given by [Eqs. (3.50) and (3.51)]

$$c_3(q) = c_3^0(g^2\mu_B^2/\chi_q^{zz})^{1/2}, c_4(q) = c_4^0 + c_4^1 g^2\mu_B^2/\chi_q^{zz},$$

$$c_s(q) = c_4^0 + \frac{(c_4^1 + \frac{1}{2}c_3^{02}/c_4^0)g^2\mu_B^2}{\chi_q^{zz}} \tag{5.4}$$

$$+ \text{higher-order terms.}$$

For the diffusion "constants" one finds in lowest order

$$\tilde{D}^{33}(q) = \tilde{d}_{33} g^2\mu_B^2/\chi_q^{zz},$$

$$\tilde{D}^{34}(q) = \tilde{d}^{34}(g^2\mu_B^2/\chi_q^{zz})^{1/2} + O((\chi_q^{zz})^{-3/2}) \; ,$$

$$\tilde{D}^{44}(q) = \tilde{d}_{44} + d'_{44} g^2\mu_B^2/\chi_q^{zz} \; . \tag{5.5}$$

Whereas the q dependence of the velocities is only a consequence of the singular susceptibility, expressions (5.5) for the transport coefficients contain the *assumption* that the Kubo formulas, or equivalently the relaxation time, are regular for $T \ll T_c$. Although very plausible, this assumption is questionable, since mode-mode coupling theory[16] gives a singularity in $\tilde{D}^{33}$ (Sec. V C). In this order it is not yet necessary to include any other q-dependent terms. Under the assumption that $\omega_P \ll c_s(q)q$ [region (i)] we find from Eq. (4.4) in lowest order in $(\chi_q^{zz})^{-1}$ the modes

$$\omega_\pm = \pm c_s(q)q - \tfrac{1}{2}i\{q^2(\tilde{d}^{44} + \tfrac{4}{3}\eta + \zeta + \zeta') + \omega_P\} \; , \tag{5.6}$$

and

$$\omega_1 = -(iq^2/c_4^{02}) \times\{\tilde{d}_{33}(c_4^0)^2 + 2\,\mathrm{Re}\,\tilde{d}^{34}c_4^0c_3^0 + \tilde{d}^{44}(c_3^0)^2\}g^2\mu_B^2/\chi_q^{zz} \; . \tag{5.7}$$

We see that there is a propagating mode with dispersion and a diffusive mode whose diffusion constant is proportional to $(\chi_q^{zz})^{-1}$ in relaxation time approximation.

In the order kept in Eqs. (5.6) and (5.7), one may still use expression (4.13) for the dynamic form factor. One only has to replace $(\partial M/\partial H)_T$ by χ_q^{zz} and c_3, c_4, c_s, D_s, D_T by the expression we have obtained now. The strength of each of the propagating poles (5.6) is (independent of the damping constants)

$$\tfrac{1}{4}T\chi_q^{zz}[c_3(q)/c_s(q)]^2 \approx \tfrac{1}{4}(g\mu_B)^2(c_3^0/c_4^0)^2 T \quad , \tag{5.8}$$

whereas the strength of the diffusive pole, which gets more and more elastic for $q \to 0$, is given by

$$\tfrac{1}{2}T\chi_q^{zz}[1 - (c_3/c_s)^2] \approx \tfrac{1}{2}[\chi_q^{zz} - g^2\mu_B^2(c_3^0/c_4^0)^2]T \, . \tag{5.9}$$

In Appendix C we have evaluated $c_3(q)$, $c_4(q)$ in the lowest-order magnon model. For $q \to 0$, $H \to 0$, χ_q^{zz} diverges, c_3 goes to zero, the strength (5.8) increases to a finite value, while (5.9) becomes infinite.

In the purely diffusive region, (iv) of Sec. IV, the diffusion constants are given also by expressions like Eq. (5.5) if one makes again the regularity assumption. We find for the modes in lowest order in $(\chi_q^{zz})^{-1}$ from Eq. (3.22),

$$\omega_\pm = -iq^2D_\pm(q) = -iq^2\{d_{44} + [(d_{34})^2/d_{44}]g^2\mu_B^2/\chi_q^{zz}\}$$

$$= -iq^2[d_{33} - (d_{34})^2/d_{44}]g^2\mu_B^2/\chi_q^{zz} \; . \tag{5.10}$$

In Sec. V C, we present the results of mode-mode theory, which are more reliable than (5.10). The large transverse fluctuations produce singularities in the Kubo integrals, which cancel the longitudinal susceptibility in D^{33}.

C. Transverse Modes

In the limit of zero external field, the equations of motion for the transverse components have also the structure of conservation laws [see Eq. (3.5)]:

$$\dot{S}^{+}_{\vec{q}} = -iq^{\alpha} j^{+\alpha}_{\vec{q}}, \quad \dot{S}^{-}_{\vec{q}} = -iq^{\alpha} j^{-\alpha}_{\vec{q}} \quad , \tag{5.11}$$

where the currents are given by

$$\begin{Bmatrix} j^{+\alpha}_{\vec{q}} \\ j^{-\alpha}_{\vec{q}} \end{Bmatrix} = -\sum_{\vec{k}} \frac{\partial \tilde{J}(\vec{k})}{\partial k_\alpha} \begin{Bmatrix} S^{z}_{(\vec{q}/2)-\vec{k}} & S^{+}_{(\vec{q}/2)+\vec{k}} \\ S^{z}_{-(\vec{q}/2)-\vec{k}} & S^{-}_{(\vec{q}/2)+\vec{k}} \end{Bmatrix} + O(q^2) . \tag{5.12}$$

We define the normalized operators

$$X^{1}_{\vec{q}} = S^{+}_{\vec{q}}/(2\chi^{T}_{q})^{1/2}, \quad X^{2}_{\vec{q}} = S^{-}_{\vec{q}}/(2\chi^{T}_{q})^{1/2} , \tag{5.13}$$

where χ^{T}_{q} is the transverse susceptibility

$$\chi^{T}_{q} = \tfrac{1}{2}\langle S^{+}_{\vec{q}} , S^{+}_{\vec{q}} \rangle . \tag{5.14}$$

The projection of $j^{+}_{\vec{q}}$ onto $P^{\alpha}_{\vec{q}}$, $M_{\vec{q}}$, $\mathcal{H}^{e}_{\vec{q}}$ vanishes. Only the scalar product

$$-iq^{\alpha}\langle j^{+\alpha}_{\vec{q}}, X^{1}_{\vec{q}} \rangle = -i\langle \textstyle\sum_{\vec{l}} S^{z}_{\vec{l}} \rangle (2\chi^{T}_{q})^{-1/2} \tag{5.15}$$

is different from zero (below T_c). Therefore the nonconserved part of the time derivatives is given by

$$q^{\alpha}\begin{Bmatrix} j^{+\alpha}_{\vec{q}} \\ j^{-\alpha}_{\vec{q}} \end{Bmatrix} + \frac{2\langle \sum S^{z}_{l} \rangle}{(2\chi^{T}_{q})^{1/2}} \begin{Bmatrix} -X^{1}_{\vec{q}} \\ X^{2}_{\vec{q}} \end{Bmatrix} \equiv \frac{2M}{(2\chi^{T}_{q})^{1/2}} \begin{Bmatrix} \tau^{+}_{\vec{q}} \\ \tau^{-}_{\vec{q}} \end{Bmatrix} . \tag{5.16}$$

Applying again the results of Sec. II, one finds

$$\frac{d}{dt}\begin{Bmatrix} \delta X^{1}_{\vec{q}} \\ \delta X^{2}_{\vec{q}} \end{Bmatrix} = i\omega(q)\begin{Bmatrix} -\delta X^{1}_{\vec{q}} \\ \delta X^{2}_{\vec{q}} \end{Bmatrix} - \Lambda(q)\begin{Bmatrix} \delta X^{1}_{\vec{q}} \\ \delta X^{2}_{\vec{q}} \end{Bmatrix} , \tag{5.17}$$

with

$$\omega(q) = \langle \textstyle\sum_{\vec{l}} S^{z}_{\vec{l}} \rangle / \chi^{T}_{q} = q^2 M_0 k_B T a^3/(M\xi\hbar) , \tag{5.18}$$

and

$$\Lambda(q) = M^2/(\chi^{T}_{q})^2$$

$$\times \tfrac{1}{2}\beta \int_{-\infty}^{\infty} dt\, e^{i\omega(q)t} \langle \tfrac{1}{2}\{\tau_{\vec{q}}(t), \tau_{\vec{q}}(0)^{\dagger}\}\rangle_c . \tag{5.19}$$

In Eq. (5.18), we have used the representation

$$\chi^{T}_{q} = M^2\xi/q^2 k_B T N a^3 (g\mu_B)^2 \tag{5.20}$$

for the transverse susceptibility. Relations (5.17) and (5.18) have already been obtained by Mori and Kawasaki.[17] Calculating the damping terms for $T \ll T_c$ self-consistently by mode-mode theory[16] one finds $D^{33} \propto q^0$, $D^{34} \propto q^{1/6} + O(q^{1/2})$, $D^{44} \propto q^0$, $\Lambda(q) \propto q^4[1 + \ln(q_m/q)]$.[19] The maximal hydrodynamic wave number is denoted by q_m. In this derivation, we factorized the correlation functions in Eqs. (3.17)–(3.19) and (5.19), inserted the hydrodynamic correlation functions, did not take $\lim_{\omega \to 0} \lim_{q \to 0}$, and replaced ω by the real part of the mode. In the regime of the second magnon, D_s and D_T are proportional to q^0. Summation of higher-order mode-mode diagrams may contribute additional logarithmic factors in $\Lambda(q)$. Recent perturbation theoretical calculations[20] gave for the damping

$$q^4 T^2[A + B\ln(T/q^2) + C\ln^2(T/q^2)] .$$

Halperin and Hohenberg[21] have predicted a q^4 damping.

From (5.17), one finds for the absorptive part of the transverse susceptibility

$$\chi''^{xx}(\vec{q}, \omega) = \tfrac{1}{2}\chi^{T}_{q}\left(\frac{\omega\Lambda(q)}{[\omega - \omega(q)]^2 + \Lambda(q)^2} + \frac{\omega\Lambda(q)}{[\omega + \omega(q)]^2 + \Lambda(q)^2}\right) . \tag{5.21}$$

The f sum rule puts a lower limit on the transverse susceptibility

$$\chi^{T}_{q} \geq M^2\left(\tfrac{1}{6} q^2 N \textstyle\sum_{\vec{l}} l^2 J(\vec{l})[g^{11}(\vec{l}) + g^{33}(\vec{l})] + HM\right)^{-1} \tag{5.22}$$

in accordance with the representation (5.20).

In the immediate vicinity of the Curie point, the hydrodynamic response functions have to be modified. Local hydrodynamics is only valid for wavelengths λ larger than the critical correlation length ξ, which increases indefinitely on approaching the transition point. For $\lambda < \xi$ a phenomenological description (dynamical scaling)[21,22] is possible, which, as static scaling, rests on the fact (or assumption) that there is only one length determining the critical behavior and that therefore susceptibilities, eigenfrequencies, etc., have to be homogeneous functions of λ and ξ. In the present paper, no discussion of the critical behavior is given.

Concluding this section we would like to elucidate shortly the absence of coupling between the equations of $M_{\vec{q}}$, $\mathcal{H}^{e}_{\vec{q}}$, $P^{\alpha}_{\vec{q}}$, and those for $S^{\pm}_{\vec{q}}$. If the Hamiltonian contained terms of the type $S^{+}S^{z}$, which, of course, would violate conservation of $\sum S^{z}_{\vec{l}}$, the transverse components would be coupled to the energy density. Another possibility is that $\sum S^{z}_{\vec{l}}$ is conserved but the ground state has different symmetry, as in a planar ferromagnet (e.g., $\langle S^{x} \rangle \neq 0$). Then the transverse components and $M_{\vec{q}}$ are coupled. Halperin and Hohenberg[23] have derived hydrodynamic equations for a planar ferromagnet in analogy with two fluid hydrodynamics.

VI. APPLICATIONS: NEUTRON AND BRILLOUIN SCATTERING

In this section, we shall determine the range of validity of this theory and the conditions for the observability of the second magnon.

A. Relaxation Rates

In order that the present theory be applicable to real systems dipole forces, interactions with phonons and impurity scattering have to be negligible. Dipole forces violate the conservation of the z component of the magnetization. If the dipole relaxation rate is much smaller than the exchange-collision rate, one may neglect dipole forces compared to the exchange forces as we have done. This requirement gives the following condition[24,25]:

$$T \gg JS(g\mu_B M_0/JS)^{4/7} . \qquad (6.1)$$

In the temperature region determined by condition (6.1), phonon-magnon processes also are much slower than the exchange processes.[24,13,26]

An average exchange collision rate is given by

$$\begin{aligned}\omega_N = {} & (2\pi/\hbar)\,[V/(2\pi)^3]^2\tfrac{1}{2}\int_{-\infty}^{\infty} d^3k_1 \cdots d^3k_4 \\ & \times [2(Ja^2/N)\vec{k}_1\cdot\vec{k}_2]^2\,\Delta(\vec{k}_1+\vec{k}_2-\vec{k}_3-\vec{k}_4) \\ & \times \delta(E_{k_1}+E_{k_2}-E_{k_3}-E_{k_4}) \\ & \times n(k_1)\,n(k_2)\,[n(k_3)+1]\,[n(k_4)+1]/\int d^3k\, n(k) ,\end{aligned} \qquad (6.2)$$

where we have extended the integration to ∞ and used the small k form of the interaction potential. The energy E_k is given by Eq. (C4) and $n(k)$ is the Bose distribution function $[\exp(\beta E_k+\alpha)-1]^{-1}$. Because of the inequality $n(k_i) > e^{-E_{k_i}/T-\alpha}$, one obtains the following lower limit for the collision frequency:

$$\omega_N > (JS/\hbar S^2)\,(T/JS)^4\,[15\,e^{-2\alpha}/2^7\sqrt{2}\,\pi^3 F_{3/2}(\alpha)] . \qquad (6.3)$$

For $\alpha=0$, one finds

$$\omega_N \gtrsim (JS/\hbar S^2)\,(T/JS)^4\, 0.001 . \qquad (6.4)$$

One obtains the same order of magnitude for the relaxation time of a thermal magnon.[20] According to Sec. IV, Eq. (4.7), we have the condition $cq \ll \omega_N$. Combining this with (6.4) and using (C6) for the sound velocity, we arrive at an upper limit for the wave number

$$aq \ll (T/JS)^{7/2}\, S^{-2}\, 10^{-3}. \qquad (6.5)$$

In order to find the condition for the existence of a frequency window (Sec. IV) we have to calculate ω_P given by (3.55). It is extremely delicate to make a reliable analytic estimate of ω_P. A possibly rough estimate gives

$$\omega_P = \gamma\,(T/JS)^{-x}\; e^{-2\omega(k_D/2)/T}\;\omega_N , \qquad (6.6)$$

where $\gamma \lesssim 1$ and x is of order 1. The vector $\vec{k}_D$ lies at the boundary of the first Brillouin zone. For fcc lattices with nearest-neighbor interaction one finds for the frequency at $\frac{1}{2}\vec{k}_D$ and for the Debye temperature

$$\omega(\tfrac{1}{2}\vec{k}_D) = 8JS, \qquad T_D=\omega(\vec{k}_D)=16JS . \qquad (6.7)$$

Here we used the full dispersion relation for a fcc lattice with nearest-neighbor interaction,[13]

$$\omega(\vec{k}) = SJ\left(12-\sum_{\delta\,\epsilon\,\mathrm{nn}} e^{i\vec{k}\cdot\vec{x}_\delta}\right) , \qquad (6.8)$$

and $\vec{k}_D=(2\pi/a, 0, 0)$. Condition (6.5) gives $qa < 10^{-3}S^{-2}$, $1.3\times10^{-1}S^{-2}$, $0.53S^{-2}$ for T equal to JS, $4JS$, and $6JS$, respectively. If, in expression (6.6) for ω_P, the factors in front of the exponential function are about 1, there is an appreciable frequency window for these temperatures. The lower limit in T set by condition (6.1) is typically of the order $10^{-1}JS$ or smaller for insulating ferromagnets. At these temperatures, the hydrodynamic q space is so small that it can scarcely be detected by experiment.[26]

It must be emphasized that for temperatures of the order of JS and higher, Eqs. (6.4) and (6.6) are no longer reliable and can at best give a very crude estimate. In place of these values, numerical evaluations of (3.55) and of the transport coefficients should be used.

Ideal insulating ferromagnetic substances are the europium chalcogenids[27] (EuO, EuS, etc.). If, with the above reservations, one applies our estimates to EuO [fcc, $a=5.14$ Å, $S=\frac{7}{2}$, $JS\approx5.4$ °K, $M_0=1910$ G, $T_c=69$ °K] one would expect to find the second magnon for temperatures 5, 20, and 30 °K, respectively, in the following q ranges:

$$[0,\ 1.6\times10^{-5}]\,,\ [4\times10^{-5},\ 2\times10^{-3}],$$
$$[5\times10^{-4},\ 0.8\times10^{-2}]\,\text{Å}^{-1}.$$

The expected velocity is $c_s\approx1.5\times10^5$ cm/sec. Impurities, dislocations, etc., can constitute a serious problem. Even elastic impurity scattering dissipates momentum and gives rise to a term $-\omega_I P_{\vec{q}}$ in the equation of motion for the momentum. The sample has to be pure enough so that $\omega_I \ll \omega_N$; then $\omega_I P_{\vec{q}}$ can be treated on the same level as $\omega_P P_{\vec{q}}$.

B. Neutron and Brillouin Scattering

An ideal probe for measuring the excitations of a magnetic system is neutron scattering. The inelastic scattering cross section for unpolarized neutrons is given by[28]

$$\frac{d\sigma}{d\Omega\,d\omega}=\frac{k_1}{k_0}\left(1.91\frac{e}{\hbar c}\right)^2\frac{1}{\pi}\left(\delta_{ij}-\frac{q_iq_j}{q^2}\right)S^{ij}(\vec{q},\omega) . \qquad (6.9)$$

The wave numbers of the incoming and outgoing neutron are k_0 and k_1, the momentum transfer is q. The dynamic form factor is defined through

$$\begin{aligned}S^{ij}(\vec{q},\omega) = {} & (g\mu_B)^2\sum_{\vec{l}}\int dt\, e^{i\omega t-i\vec{q}\cdot\vec{x}_{\vec{l}}} \\ & \times\tfrac{1}{2}\langle\langle(S^i_{\vec{l}}-\langle S^i_{\vec{l}}\rangle)\,(S^j_{\vec{0}}-\langle S^j_{\vec{0}}\rangle)\rangle\rangle .\end{aligned} \qquad (6.10)$$

It is related to the imaginary part of the dynamic susceptibility by the fluctuation dissipation theorem:

$$S^{ij}(\vec{q},\omega) = (1-e^{-\beta\omega})^{-1}\chi''^{ij}(\vec{q},\omega) \approx (T/\omega)\,\chi''^{ij}(\vec{q},\omega)\,. \tag{6.11}$$

The second identity is valid in the hydrodynamic regime.

As is seen from (6.9) the scattering cross section is a superposition of transverse $S^{xx}(\vec{q},\omega) = S^{yy}(\vec{q},\omega)$ and longitudinal $S^{zz}(\vec{q},\omega)$ dynamic form factors. If the window condition is met, the longitudinal form factor contains the second magnon and a diffusive mode, and is given by Eq. (4.13). In the opposite case if $cq \lesssim \omega_P$, $S^{zz}(\vec{q},\omega)$ is purely diffusive [regions (ii–iv) of Sec. IV]. In a perfect single crystal without domains one could isolate $S^{zz}(\vec{q},\omega)$ by measuring $d\sigma/d\Omega\, d\omega$ for $\vec{q}$ in the [1, 0, 0] and [0, 0, 1] directions and subtracting half of the latter from the former. By applying an external field, one could eliminate domains and shift the transverse modes to higher frequency.

At this point we want to discuss the effects of an external magnetic field. First, it reduces ω_N and the hydrodynamic q space, because the population of thermal magnons becomes smaller. This effect makes itself felt for $H > k_B T/g\mu_B$. Second, according to the estimates of Appendix C the relative strength of propagating modes to diffusive mode increases. Third, the static longitudinal susceptibility decreases, which enters as a factor in $S^{zz}(\vec{q},\omega)$. If one applies the simple magnon model of Appendix C one finds an acceptable value for the field given by $10^{-1} \lesssim g\mu_B H/k_B T \lesssim 1$, which at 20 °K gives $10^4\,\mathrm{G} \lesssim H \lesssim 10^5\,\mathrm{G}$. For comparison with experiment more realistic values for the thermodynamic derivatives should be taken.

A further experimental check of the present theory could be possible with Brillouin scattering. For this purpose one needs a transparent ferromagnet, an example of which is[29] $\mathrm{Eu_2SiO_4}$. Also $\mathrm{CrBr_3}$ transmits light in two bands, one in the red and one in the green.[30] This uniaxial ferromagnet crystallizes in a hexagonal layer structure. Using the values[31]

$$J_1 S = (1.491 \pm 0.039)\,^\circ\mathrm{K}, \quad J_t S = (24.75 \pm 0.3)\,^\circ\mathrm{K}\,,$$

one sees that the surface of constant spin-wave energy is an ellipsoid which is very prolate along c and which touches the zone boundaries for energies of about 5 °K. For higher energies the surfaces of constant energy are cylinders. The momentum relaxation rate $\omega_P^{\alpha\beta}$ is highly anisotropic. There are many umklapp processes in the c direction but the relaxation rate for momentum orthogonal to the c axis $\omega_P^{\perp}$ will still be small for a temperature of 10 to 15 °K.

Besides the high purity required of the sample, the main difficulty for an observation by neutron scattering is at present time the low momentum transfer. One may hope that this will be accessible to high-flux reactors.

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APPENDIX A: THERMODYNAMIC RELATIONS

Here we want to compile thermodynamic relations needed in the main text. The thermodynamics of the ferromagnet is generalized so as to treat the volume as a thermodynamic variable. It is shown that the pressure defined through the trace of the momentum-flux operator is identical with the volume derivative of the internal energy, neglecting terms of the order $e^{-T_D/T}$.[32]

The density matrix of the system is

$$\rho = Z^{-1}\exp[-\beta(\mathcal{H}^e - Hg\mu_B \textstyle\sum_{\vec{l}} S_{\vec{l}}^z)]\,, \tag{A1}$$

where Z is the partition function

$$Z = e^{-\beta F} = \mathrm{Tr}(\exp[-\beta(\mathcal{H}^e - Hg\mu_B \Sigma S_{\vec{l}}^z)])\,. \tag{A2}$$

If there are single-ion terms or any other internal interactions which do not violate the conservation of $\Sigma S_{\vec{l}}^z$, we include them in $\mathcal{H}^e$. The thermal expectation value of any operator O is given by

$$\langle O \rangle = \mathrm{Tr}(\rho O)\,. \tag{A3}$$

The internal energy, excluding the interaction with the external field,

$$U = \langle \mathcal{H}^e \rangle\,, \tag{A4}$$

and the magnetization in the z direction,

$$M = g\mu_B \textstyle\sum_{\vec{l}} \langle S_{\vec{l}}^z \rangle\,, \tag{A5}$$

is related to the entropy by

$$TS \equiv -T\langle \ln\rho \rangle = -F + U - HM\,. \tag{A6}$$

We also note the differential relation

$$dU = T dS + H dM \equiv T dS + H d(M - M_0)\,. \tag{A7}$$

Differentiating (A4) and (A5) gives for the $q = 0$ static susceptibilites

$$\langle \mathcal{H}_0^e, \mathcal{H}_0^e \rangle = T\left(\frac{\partial U}{\partial T}\right)_H + H\left(\frac{\partial U}{\partial H}\right)_T\,, \tag{A8}$$

$$\langle \mathcal{H}_0^e, M_0 \rangle = \left(\frac{\partial U}{\partial H}\right)_T = T\left(\frac{\partial M}{\partial T}\right)_H + H\left(\frac{\partial M}{\partial H}\right)_T\,, \tag{A9}$$

$$\langle M_0, M_0 \rangle = \left(\frac{\partial M}{\partial H}\right)_T\,. \tag{A10}$$

For variations of long wavelength such that the system is in local equilibrium, the fluctuations in energy, magnetization, temperature, and field are related through

$$\delta U(\vec{q}) = \left(\frac{\partial U}{\partial \beta}\right)_H \delta\beta(\vec{q}) + \left(\frac{\partial U}{\partial H}\right)_\beta \delta H(\vec{q})\,,$$

$$\delta M(\vec{q}) = \left(\frac{\partial M}{\partial \beta}\right)_H \delta\beta(\vec{q}) + \left(\frac{\partial M}{\partial H}\right)_\beta \delta H(\vec{q}) \,. \tag{A11}$$

This implies

$$\delta\beta(\vec{q}) = \left[\left(\frac{\partial M}{\partial H}\right)_\beta \delta U(\vec{q}) - \left(\frac{\partial U}{\partial H}\right)_\beta \delta M(\vec{q})\right]\left(\frac{\partial (U, M)}{\partial (\beta, H)}\right)^{-1} . \tag{A12}$$

Application of the identity

$$\frac{\partial (U, M)}{\partial (\beta, H)} = T(-\langle \mathcal{H}_0^e,\ \mathcal{H}_0^e\rangle \langle M_0,\ M_0\rangle + \langle \mathcal{H}_0^e,\ M_0\rangle^2)$$

$$= -T^2 C_M\left(\frac{\partial M}{\partial H}\right)_T , \tag{A13}$$

where $C_M = T(\partial S | \partial T)_M$ is the total specific heat at constant magnetization, gives for the normalization factor in (3.13)

$$d(0) = \left[T C_M\left(\frac{\partial M}{\partial H}\right)_T^2\right]^{1/2} \quad . \tag{A14}$$

Comparison of Eqs. (A12) and (3.13) shows that the nonequilibrium expectation value of $X^4_{\vec{q}}$ is related to the variation of the normalized temperature

$$\delta X^4_{\vec{q}} \equiv \langle X^4_{\vec{q}}\rangle_{\text{Nonequ.}} - \langle X^4_{\vec{q}}\rangle$$

$$= -(T^3 C_M)^{1/2}\, \delta\beta(\vec{q}) = (C_M/T)^{1/2}\, \delta T(\vec{q}) \,. \tag{A15}$$

The Hamiltonian of the ferromagnet contains a parameter, the volume V. So far we tacitly kept this parameter constant. However, it is useful to take V as a thermodynamic variable and to introduce the conjugate force, the pressure

$$\mathcal{P} = -\left(\frac{\partial U}{\partial V}\right)_{M-M_0,\, S} \quad . \tag{A16}$$

Thus, we have

$$dU = T dS - \mathcal{P}\, dV + H d(M - M_0) \quad . \tag{A7$'$}$$

As usual one can derive the Duhem-Gibbs identity

$$U = S\,T - \mathcal{P} V + H(M - M_0) \,. \tag{A17}$$

We shall now demonstrate that the expectation value of the momentum flux tensor PV [Eq. (3.40)] is identical to the thermodynamic pressure [Eq. (A16)] neglecting terms of the order $e^{-T_D/T}$. First of all we note the identity

$$\mathcal{P} = -\frac{\partial\Omega}{\partial N_\alpha a} = -\left\langle\frac{\partial(\mathcal{H}^e + g\,\mu_B M_0)}{\partial N_\alpha a}\right\rangle \quad . \tag{A18}$$

The volume change is to be understood as a change of N_α; the lattice constant a remains constant. The Hamiltonian (3.33) depends on N_α through the factor $1/N$ and through the Fourier-transformed exchange constant which we write explicitly as

$$\tilde{J}(\vec{k}) \equiv \tilde{J}(g_\alpha \nu_\alpha / N_\alpha) \,.$$

The operators $a_{\vec{k}} \equiv a_{\vec{\nu}}$, of course, contain no N-dependence. The summation over ν_α extends from $-\frac{1}{2}N_\alpha$ to $\frac{1}{2}N_\alpha$. For low temperatures one may extend the summation from $-\infty$ to $+\infty$, committing an error of order $e^{-T_D/T}$. Employing Eq. (A18) one finds by straightforward differentiation

$$P = \mathcal{P} + O(e^{-T_D/T}) \,. \tag{A19}$$

Using (A8 – A10), (A14), (A17), and (A19), expression (3.51) for c_4 may be written in the thermodynamic limit as

$$c_4 = (V\rho\, T C_{M,V})^{-1/2}\, T\left[S - (M_0 - M)\left(\frac{\partial H}{\partial T}\right)_{M-M_0,\, V}\right],$$

and from the Duhem-Gibbs identity (A17) we get

$$c_4 = \left(\frac{\rho}{V}\ C_{M,V}\right)^{-1/2} (\sqrt{T})\left(\frac{\partial P}{\partial T}\right)_{M,V} \quad . \tag{A20}$$

Similarly, one finds

$$c_3 = \left[-\frac{\rho}{V}\left(\frac{\partial V}{\partial P}\right)_{T,\, M-M_0}\right]^{-1/2} . \tag{A21}$$

and for $$c_s = \left[-\frac{\rho}{V}\left(\frac{\partial V}{\partial P}\right)_{S,\, M-M_0}\right]^{-1/2} \quad . \tag{A22}$$

APPENDIX B: ANISOTROPIC EXCHANGE INTERACTION

We quote the modifications which arise from an anisotropic exchange interaction. We assume that the Hamiltonian (3.2) contains an additional term,

$$\Delta\mathcal{H} = -\tfrac{1}{2}\sum_{l,l'} \Delta\tilde{J}_\parallel(\vec{l} - \vec{l}\,')\, S^z_l\, S^z_{l'} \,, \tag{B1}$$

where $\Delta\tilde{J}_\parallel(\vec{l} - \vec{l})$ is such that it favors ferromagnetic ordering in the z direction. One finds the following additional terms to the currents:

$$\Delta j^{M\alpha}_{\vec{q}} = 0 \,, \tag{B2}$$

$$\Delta j^{E\alpha}_{\vec{q}} = \frac{1}{2}\sum_{\vec{l},\vec{l}',\vec{l}''} e^{-(i/2)\vec{q}\cdot(\vec{X}_{\vec{l}} + \vec{X}_{\vec{l}''})}$$

$$\times (X_{\vec{l}} - X_{\vec{l}''})^\alpha\, \Delta J_\parallel(\vec{l}'' - \vec{l}')\, J(\vec{l} - \vec{l}')$$

$$\times(-S^z_{\vec{l}''} S^x_{\vec{l}}\, S^y_{\vec{l}'} + S^z_{\vec{l}''} S^y_{\vec{l}}\, S^x_{\vec{l}'}) \,, \tag{B3}$$

$$\Delta\Pi^{\alpha\beta}_{\vec{q}} = -\frac{1}{2}\sum_{\vec{k}_1,\dots\vec{k}_4} (k_1 - k_4)^\alpha \frac{\partial}{\partial k^\beta}\, \Delta\tilde{J}_\parallel(\vec{k}_1 - \vec{k}_4)$$

$$\times\Delta(\vec{q} + \vec{k}_1 + \vec{k}_2 - \vec{k}_3 - \vec{k}_4)\, a^\dagger_{\vec{k}_1} a^\dagger_{\vec{k}_2} a_{\vec{k}_3} a_{\vec{k}_4} \,, \tag{B4}$$

$$\Delta u^\alpha_{\vec{q}} = -\frac{1}{2}\sum_{\vec{k}_1,\dots\vec{k}_4} \Delta\tilde{J}_\parallel(\vec{k}_1 - \vec{k}_4)\, \Delta(\vec{q} + \vec{k}_1 + \vec{k}_2 - \vec{k}_3 - \vec{k}_4)$$

$$\times g^\alpha(\vec{k}_1 + \vec{k}_2 - \vec{k}_3 - \vec{k}_4 + \vec{q})\, a^\dagger_{\vec{k}_1} a^\dagger_{\vec{k}_2} a_{\vec{k}_3} a_{\vec{k}_4} \,. \tag{B5}$$

The decomposition of the currents in its conserved and in its nonconserved parts is still given by (3.46), (3.47), and (3.49). The structure of the hydrodynamic equations remains unchanged but in the calculation of the coefficients the new thermal expectation values and the new currents have to be taken.

APPENDIX C: LOWEST-ORDER MAGNON MODEL

For a Heisenberg model with nearest-neighbor interaction one may use Dyson's[4] expression for the free energy [Eq. (131) of second paper] to find the velocities c_3, c_4 in the hydrodynamic equations. For more complex situations an empirical expression for the free energy could be used to find the necessary thermodynmaic derivatives. In order to demonstrate the principal features we take only the lowest-order (Bloch) term of Dyson's expansion. The free energy for a cubic system with exchange constant J then reads

$$\begin{aligned} F &= -\,[VT^{5/2}/(JSa^2)^{3/2}\, 8\pi^{3/2}]\, F_{5/2}(\alpha) - HM_0 \\ &= \Omega - HM_0 , \end{aligned} \tag{C1}$$

where $F_\sigma(\alpha)$ is the generalized ζ function

$$F_\sigma(\alpha) = [1/\Gamma(\sigma)] \int_0^\infty dx\, [x^{\sigma-1}/(e^{x+\alpha}-1)] , \tag{C2}$$

and α is the reduced magnetic field

$$\alpha = Hg\mu_B/T \;. \tag{C3}$$

The quantity Ω is just the Gibbs potential of an ideal Bose gas with energy spectrum

$$E_k = JS(ka)^2 \equiv \mathfrak{D}k^2 \equiv (\hbar k)^2/2m \;. \tag{C4}$$

Only the momentum fluctuations can not be derived from F. For an ideal magnon gas one readily finds

$$V\rho\,\delta^{\alpha\beta} = \langle P^\alpha_{\vec q},\; P^\beta_{\vec q}\rangle = m(M_0 - M)\,\delta^{\alpha\beta} \;. \tag{C5}$$

Herewith we get for the velocities (4.11) or (4.13)

$$\begin{aligned} c_3 &= [T F_{3/2}(\alpha)/m F_{1/2}(\alpha)]^{1/2}, \\ c_s &= [T\, 5 F_{5/2}(\alpha)/m\, 3 F_{3/2}(\alpha)]^{1/2} \;. \end{aligned} \tag{C6}$$

In this approximation the residue of each one of the propagating poles in $S^{zz}(q,\omega)$ is

$$\begin{aligned} R_\pm &= \tfrac{1}{2} T(g\mu_B)^2 (V T^{1/2}/8\pi^{3/2}\mathfrak{D}^{3/2}) \\ &\quad\times (3F^2_{3/2}/10F_{5/2}) , \end{aligned} \tag{C7}$$

and of the diffusive pole,

$$R_1 = \tfrac{1}{2} T(g\mu_B)^2\, (VT^{1/2}/8\pi^{3/2}\mathfrak{D}^{3/2})\, (F_{1/2} - \tfrac{3}{5} F^2_{3/2}/F_{5/2}) \,. \tag{C8}$$

In order to get some estimate for the diffusion constants, we evaluate the Kubo formulas by a relaxation time approximation for the currents

$$\tilde{j}^\alpha_{\vec q}(t) = \tilde{j}^\alpha_{\vec q}(0)\, e^{-|t|/\tau}, \quad \tau = \omega_N^{-1}, \tag{C9}$$

which implies that

$$\begin{aligned} &\lim \tfrac{1}{2}\beta \int_{-\infty}^{\infty} dt \,\langle \tfrac{1}{2}\{\tilde{j}^\alpha_{\vec q}(t),\; \tilde{j}^{\beta\dagger}_{\vec q}(0)\}\rangle_c\, e^{i\omega t} \\ &\quad = \beta\tau\langle \tfrac{1}{2}\{\tilde{j}^{\,\alpha}(0),\; \tilde{j}^{\beta}(0)\}\rangle_c \,. \end{aligned} \tag{C10}$$

In the equal-time correlation functions (C10) only the quadratic contributions to the currents are retained, and the resulting four-point functions are factorized. The constant given by the equal-time correlation function yields only an order-of-magnitude estimate. In the case of a gas these approximations reproduce the results of the kinetic theory. One finds for the thermal diffusivity and the viscosity

$$\begin{aligned} D^{44} &= (T\tau/m)\tfrac{5}{3}\,(7F_{7/2} - 5F^2_{5/2}/F_{3/2}) \\ &\quad \times (5F_{5/2} - 3F^2_{3/2}/F_{1/2})^{-1}, \\ \eta &= [(T\tau/m)F_{5/2}]/F_{3/2} \,. \end{aligned} \tag{C11}$$

The other diffusion constants are different from zero only if one includes interaction or nonquadratic terms in the dispersion.

We calculate now the q-dependent sound velocities of Sec. V [Eq. (5.4)]. Using (5.2), (C1), and (C5) in Eqs. (3.50) and (3.51) we find

$$\begin{aligned} c_3(q) &= [T F_{3/2}(\alpha)/m\psi\pi^{3/2}]^{1/2}\, (\mathfrak{D}q^2/T)^{1/4} + O(q^{3/2}) , \\ c_4(q) &= [T\, 5F_{5/2}(\alpha)/m\, 3F_{3/2}(\alpha)]^{1/2} \\ &\quad \times \{1 - [3F^2_{3/2}(\alpha)/10\psi\pi^{3/2}F_{5/2}(\alpha)] \\ &\quad \times (\mathfrak{D}q^2/T)^{1/2}\} + O(q^2) , \\ c_s(q) &= [T\, 5F_{5/2}(\alpha)/m\, 3F_{3/2}(\alpha)]^{1/2} + O(q^2) \;. \end{aligned} \tag{C12}$$

From these values in conjunction with Eq. (5.2) we obtain for the strength of each one of the propagating poles in $S^{zz}(q,\omega)$ again $R_\pm$ given in Eq. (C7). The strength of the diffusive pole is $R_1 = \frac{1}{2} T \chi^{zz}_q - -\frac{1}{2}R_+$.

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Raman Spectra and Mode Frequency Shifts of Ferroelectric Sodium Nitrite at 77 and 294°K

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The Raman spectra of the noncentrosymmetric biaxial sodium nitrite ($NaNO_2$) single crystal at 77 and 294 °K are presented. No major difference between the two spectra is observed, indicating that the recently reported phase transition at 178 °K from thermal-expansion measurements does not appear to change the crystal symmetry. Frequency shifts were observed with changes in the phonon wave-vector orientation for all five asymmetric B modes. The frequency shifts of the asymmetric internal vibrational $B_1(x)$ mode of the NO_2^- molecular ion were examined in some detail.

INTRODUCTION

Very little work has been done on the study of noncentrosymmetric biaxial crystals by Raman spectroscopy. The lack of theory, as well as the birefringence, makes the analysis of the data difficult at best. Ferroelectric sodium nitrite is one of the simplest orthorhombic biaxial crystals consisting of one molecule, four atoms, per primitive unit cell. The limited number of normal modes