

Thermodynamic approach in the theory of paramagnetic resonance of magnons

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The possibility is demonstrated of thermodynamically describing quasistationary distributions of magnons excited parametrically by powerful microwave pumping pulses. The basic idea is that the system Hamiltonian is stationary in a rotating coordinate frame and the problem reduces to that of perturbed Bose-quasiparticle gas that approaches thermodynamic equilibrium with a certain effective temperature. The basic equations for the resultant states are derived and solved exactly. Equations are obtained for the effective temperature in the cases of instantaneously and adiabatically applied pump fields. Criteria for thermodynamic stability of the states are considered. The spectrum of the collective oscillations of the system is analyzed. An interpretation is proposed for some experimentally observed effects.

INTRODUCTION

The spin-wave concept introduced in physics by Bloch more than fifty years ago explains many thermodynamic and kinetic phenomena in magnetically ordered systems. By using this concept, perturbations of a magnetic material can be regarded as an evolution of a nonideal gas of quasiparticles (magnons) or of an interaction, in a nonlinear medium, of waves excited to some definite level. Thus, investigations of various (not necessarily small) perturbations in magnetic systems permit different processes in nonlinear multiparticle media to be studied and simulated.

A most convenient method of exciting a large (macroscopic) number of magnons in a ferro- or antiferromagnetic sample is parametric resonance of spin waves in a microwave field $h \cos \omega_p t$ (Refs. 1–5). In the case of degenerate pumping, a microwave-field quantum decays in a magnetic medium into two magnons having half the frequency and oppositely directed wave vectors: $\omega_p = \omega_k + \omega_{-k}$.

Magnons are parametrically excited only when a threshold field amplitude h_c is exceeded, in which case the number of magnons begins to increase exponentially with a growth rate $\sim h/h_c - 1$. This magnon generation can lead to fundamentally different end results, depending on whether the magnetic system is or is not in contact with a heat bath (thermostat). In the former case, nonlinear interactions stabilize the energy flow from the pump to the excited magnons that relax subsequently to form the thermostat (other magnons, phonons etc.). There are two known independent mechanisms for this stabilization: 1) positive nonlinear damping,^{6,7} wherein the relaxation rate of the parametric magnons increases as their number increases, and 2) the so-called phase mechanism,⁸ wherein the forced oscillations of the magnetic medium, of frequency $\omega_p/2$, deviate in phase from the pump field. Obviously, the flows are theoretically in equilibrium if the thermostat is infinitely large. Principal attention was in fact paid initially in magnon-parametric-resonance theory to the equilibrium states of the fluxes, and definite advances have been made in the description of the evolution of excited slightly supercritical systems with $h/h_c - 1 \lesssim 1$ (Refs. 8–18).

Parametrically excited magnons can evolve to an entirely different state if a finite reservoir is involved, e.g., if the magnons are isolated from other degrees of freedom of the crystal. Collisions between quasiparticles lead to redistribu-

tion of the absorbed energy in the magnon gas, and this helps establish a steady-state nonequilibrium distribution in the gas after a certain time τ_m (which depends on the magnon density). This means that the excited magnon system saturates and ceases to absorb energy from the pump field. For real objects, the approximation of an isolated magnon system is tenable, obviously, only if within a finite time interval $\tau \ll \tau_{ph}$, where τ_{ph}^{-1} is the characteristic rate at which the excited magnons relax to form a phonon system. This evolution of parametrically excited magnons can therefore be observed in experiment by using a pulse technique with pulse length τ , where $\tau_m \ll \tau \ll \tau_{ph}$. This inequality can be satisfied for a large number of magnets, in view of the different dependence of τ_m and τ_{ph} on the density N of the excited magnons, N : $\tau_m/\tau_{ph} \sim N^{-2}$ and also because magnon–magnon anharmonicities are much more effective than magnon–phonon nonlinear interactions. An example of a system in which this inequality can be satisfied with sufficient margin is a system of nuclear spin waves in an antiferromagnet, for which the spin–lattice times at a temperature of order 10^{-2} K reach several days.¹⁹ Pulse investigations provide information only on interactions inside the magnon system. In the present paper we develop a theory for the states for a states of a parametrically excited magnon system that has reached saturation in a pump field.

The saturation effect is easiest to understand by using a coordinate frame that “rotates” at a frequency $\omega_p/2$. In this frame the magnetic Hamiltonian of interest to us does not depend explicitly on the time. This Hamiltonian can be reduced by a Bogolyubov canonical (u, v) transformation²⁰ to a form that describes a weakly nonideal Bose gas of quasiparticles. Henceforth we shall use the term “quasiparticle” for just these excitations. Turning on the microwave pump field means creation of a definite number of quasiparticles whose mutual scattering leads to a thermodynamic equilibrium at a certain temperature Θ . It should be noted that the idea of a thermodynamic description of an object in a rotating reference frame as applied to NMR was first set forth by Redfield (see Refs. 21 and 22). In recent theoretical papers^{23,24} the thermodynamic approach in a rotating frame is used also for systems with a continuous spectrum, such as the Fermi system of the semiconductor in a laser-pulse field.

In the present paper we use the thermodynamic approach in a rotating frame to describe a Bose system. Note that a change to representation of quasiparticles in a rotating

coordinate frame was used earlier in Refs. 11 and 13, but in connection with the evolution of parametrically excited magnons in contact with a thermostat. In addition, the results of Refs. 11 and 13 are valid only for a quasiparticle spectrum $\Omega_{\mathbf{k}}$ that is positive-definite (for all values of \mathbf{k}), and the theory developed there has a substantially narrower range of validity. We shall show below that in exactly solvable models the quasiparticle spectrum contains an interval of negative values, but this circumstance does not prevent a thermodynamic description of a thermally insulated system.

The plan of the paper is the following. In Sec. 1 we obtain the conditions for diagonalizing the magnon Hamiltonian in a rotating coordinate frame, and in Sec. 2 the realization of these conditions is demonstrated with exactly solvable cases as examples. In Sec. 3 we determine the effective temperature of a quasiparticle gas whose thermodynamic stability we verify. Equations for the spectrum of the collective excitations of the system are derived in Sec. 4. We conclude with a discussion of the results and with their relation to experiment.

1. BASIC EQUATIONS

We express the initial Hamiltonian of a magnon system in a microwave-pump field in the form

$$\begin{aligned} \mathcal{H} &= \sum_{\mathbf{k}} \left[\varepsilon_{\mathbf{k}} a_{\mathbf{k}}^+ a_{\mathbf{k}} + \frac{1}{2} h V_{\mathbf{k}} (e^{i\omega_p t} a_{\mathbf{k}} a_{-\mathbf{k}} + \text{h.c.}) \right] + \mathcal{H}_{int}, \\ \mathcal{H}_{int} &= \frac{1}{2} \sum_{1,2,3,4} \Phi(1, 2; 3, 4) a_1^+ a_2^+ a_3 a_4 \Delta(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 - \mathbf{k}_4), \end{aligned} \quad (1)$$

where $\varepsilon_{\mathbf{k}}$ is the magnon spectrum; $a_{\mathbf{k}}^+$ and $a_{\mathbf{k}}$ are Bose creation and annihilation operators; $V_{\mathbf{k}}$ is the coupling coefficient of the magnons and the pump field; $\Phi(1, 2; 3, 4)$ is the interaction matrix element. We use a system of units in which $\hbar = 1$ and $k_B = 1$.

The evolution of the system in time is described by the equation for the density matrix ρ :

$$i \partial \rho / \partial t = [\mathcal{H}, \rho].$$

Applying the unitary transformation¹¹

$$\bar{\rho} = U^{-1} \rho U, \quad U = \exp \left[-\frac{i\omega_p t}{2} \sum_{\mathbf{k}} a_{\mathbf{k}}^+ a_{\mathbf{k}} \right],$$

we obtain a Hamiltonian $\tilde{\mathcal{H}}$ that does not depend explicitly on the time:

$$\tilde{\mathcal{H}} = \sum_{\mathbf{k}} \left[\left(\varepsilon_{\mathbf{k}} - \frac{\omega_p}{2} \right) a_{\mathbf{k}}^+ a_{\mathbf{k}} + \frac{1}{2} h V_{\mathbf{k}} (a_{\mathbf{k}} a_{-\mathbf{k}} + a_{\mathbf{k}}^+ a_{-\mathbf{k}}^+) \right] + \mathcal{H}_{int}. \quad (2)$$

Next we diagonalize $\tilde{\mathcal{H}}$ by using the Bogolyubov (u, v) transformation for a Bose system:

$$a_{\mathbf{k}} = u_{\mathbf{k}} b_{\mathbf{k}} + v_{\mathbf{k}} b_{-\mathbf{k}}^+, \quad a_{\mathbf{k}}^+ = u_{\mathbf{k}} b_{\mathbf{k}}^+ + v_{\mathbf{k}} b_{-\mathbf{k}}, \quad (3)$$

$$u_{\mathbf{k}}^2 - v_{\mathbf{k}}^2 = 1. \quad (4)$$

We introduce the vacuum-state vector $|0\rangle: b_{\mathbf{k}} |0\rangle = 0$ and $\langle 0| b_{\mathbf{k}}^+ = 0$. The transformed Hamiltonian (2) averaged

over the vacuum state is then

$$\mathcal{E}_0 = \langle 0 | \tilde{\mathcal{H}} | 0 \rangle = \frac{1}{2} \sum_{\mathbf{k}} \left[\left(\varepsilon_{\mathbf{k}} - \frac{\omega_p}{2} + E_{\mathbf{k}} \right) N_{\mathbf{k}} + (h V_{\mathbf{k}} + \Delta_{\mathbf{k}}) \sigma_{\mathbf{k}} \right]. \quad (5)$$

We use for convenience the notation

$$N_{\mathbf{k}} = \langle 0 | a_{\mathbf{k}}^+ a_{\mathbf{k}} | 0 \rangle = v_{\mathbf{k}}^2, \quad (6a)$$

$$\sigma_{\mathbf{k}} = \langle 0 | a_{\mathbf{k}} a_{-\mathbf{k}} | 0 \rangle = u_{\mathbf{k}} v_{\mathbf{k}}, \quad (6b)$$

$$E_{\mathbf{k}} = \varepsilon_{\mathbf{k}} - \frac{\omega_p}{2} + 2 \sum_{\mathbf{q}} T_{\mathbf{k}\mathbf{q}} N_{\mathbf{q}}, \quad T_{\mathbf{k}\mathbf{q}} = \Phi(\mathbf{k}, \mathbf{q}; \mathbf{k}, \mathbf{q}), \quad (7a)$$

$$\Delta_{\mathbf{k}} = h V_{\mathbf{k}} + \sum_{\mathbf{q}} S_{\mathbf{k}\mathbf{q}} \sigma_{\mathbf{q}}, \quad S_{\mathbf{k}\mathbf{q}} = \Phi(\mathbf{k}, -\mathbf{k}; \mathbf{q}, -\mathbf{q}). \quad (7b)$$

In this notation Eq. (4) takes the form

$$\sigma_{\mathbf{k}}^2 = N_{\mathbf{k}}^2 + N_{\mathbf{k}}. \quad (8)$$

Minimizing (5) with account taken of (8), we obtain

$$E_{\mathbf{k}} \sigma_{\mathbf{k}} + (N_{\mathbf{k}} + 1/2) \Delta_{\mathbf{k}} = 0. \quad (9)$$

The functions $N_{\mathbf{k}}$ and $\sigma_{\mathbf{k}}$ that satisfy (8) and (9) describe the excited-magnon distribution calculated from the vacuum state of the quasiparticle Hamiltonian:

$$\tilde{\mathcal{H}} = \mathcal{E}_0 + \sum_{\mathbf{k}} \Omega_{\mathbf{k}} b_{\mathbf{k}}^+ b_{\mathbf{k}} + \tilde{\mathcal{H}}^{(4)}. \quad (10)$$

Here

$$\Omega_{\mathbf{k}} = (E_{\mathbf{k}}^2 - \Delta_{\mathbf{k}}^2)^{1/2} \text{sign}(E_{\mathbf{k}}) \quad (11)$$

is the quasiparticle frequency and $\tilde{\mathcal{H}}^{(4)}$ is a four-particle interaction Hamiltonian in which the products of the operators $b_{\mathbf{k}}^+$ and $b_{\mathbf{k}}$ are so ordered that the creation operators are on the left of the annihilation operators.

It is easy to obtain from (8) and (9) the condition under which the quasiparticle $\Omega_{\mathbf{k}}$ is real:

$$E_{\mathbf{k}}^2 - \Delta_{\mathbf{k}}^2 = \frac{1}{4} \frac{\Delta_{\mathbf{k}}^2}{N_{\mathbf{k}}^2 + N_{\mathbf{k}}} \geq 0.$$

It can be seen that it is valid for solutions of physical interest, with $N_{\mathbf{k}} \geq 0$. Note also that the alternation of the sign of the spectrum does not effect the dynamic part of the problem and is compensated for in the thermodynamic description by the chemical potential μ .

The steady-state temperature Θ of the quasiparticles depends in general on the initial conditions (this question is dealt with in detail in Sec. 3). It may turn out that no vacuum state is realized ($\Theta \neq 0$) under any physical assumption. It is necessary therefore to generalize for the case $\Theta \neq 0$ Eqs. (8) and (9) for the coefficients (3) of the conversion that reduces the Hamiltonian (2) to normal modes. Such a generalization is possible in the framework of the self-consistent-field approximation if the quasiparticle occupation numbers are small. Following Ref. 25, where a similar problem is solved for a Fermi system, we introduce the density matrix

$$\rho_0 = \frac{e^Q}{\text{Sp}[e^Q]}, \quad Q = -\frac{1}{\Theta} \sum_{\mathbf{k}} (\Omega_{\mathbf{k}} - \mu) b_{\mathbf{k}}^+ b_{\mathbf{k}}.$$

Let us determine the averages $\langle a_k^+ a_k \rangle_0$ and $\langle a_k a_{-k} \rangle_0$ over the density matrix ρ_0 with allowance for relations (3); then

$$N_k \equiv \langle a_k^+ a_k \rangle_0 = u_k^2 n_k + (n_k + 1) v_k^2, \quad (12a)$$

$$\sigma_k \equiv \langle a_k a_{-k} \rangle_0 = u_k v_k (1 + 2n_k), \quad (12b)$$

where $n_k \equiv \langle b_k^+ b_k \rangle_0$ are the quasiparticle occupation numbers, equal at the temperature Θ to

$$n_k = \{ \exp [(\Omega_k - \mu) / \Theta] - 1 \}^{-1}. \quad (13)$$

Using Wick's theorem for fourth-order correlators, we obtain now the conditions for the minimum of $\langle \mathcal{H} \rangle_0$ with a specified number n_k . We get ultimately

$$\sigma_k^2 = (N_k + 1/2)^2 - (n_k + 1/2)^2, \quad (14a)$$

$$E_k \sigma_k + (N_k + 1/2) \Delta_k = 0, \quad (14b)$$

where E_k and Δ_k are defined by expressions (7) in which N_k and σ_k are taken from (12).

The only parameter still undetermined in Eqs. (14) is the chemical potential μ ,¹⁾ which enters explicitly in expression (13) for n_k . Its value in the ideal-gas approximation with a variable quasiparticle number is, for a given energy and under conditions of maximum entropy,

$$\mu = \min_k (\Omega_k). \quad (15)$$

For a monotonically increasing function $\Omega_k = \Omega(|\mathbf{k}|)$, for example, we have $\mu = \Omega_0$.

2. EXACT SOLUTIONS

It follows from Eq. (14b) that in the quasiparticle k -space one can distinguish a characteristic region, a resonance surface with $\mathbf{k}_0 \in \mathbf{K}_0$, on which

$$E_{\mathbf{k}_0} = 0, \quad \Delta_{\mathbf{k}_0} = 0, \quad (16)$$

while outside of its Eqs. (14) can be transformed into

$$\sigma_k = -(N_k + 1/2) \Delta_k / E_k, \quad (17a)$$

$$N_k = \frac{1}{2} \left\{ \frac{2n_k + 1}{[1 - (\Delta_k / E_k)^2]^{1/2}} - 1 \right\}. \quad (17b)$$

The expressions for E_k and Δ_k can be written, with allowance for (16), in the form

$$E_k = \varepsilon_k - \varepsilon_{\mathbf{k}_0} + 2 \sum_{\mathbf{q}} (T_{\mathbf{k}\mathbf{q}} - T_{\mathbf{k}_0\mathbf{q}}) N_{\mathbf{q}}, \quad (18a)$$

$$\Delta_k = \hbar(V_k - V_{\mathbf{k}_0}) + \sum_{\mathbf{q}} (S_{\mathbf{k}\mathbf{q}} - S_{\mathbf{k}_0\mathbf{q}}) \sigma_{\mathbf{q}}. \quad (18b)$$

Clearly, the system (17) can be solved in the general case by iteration, substituting in it relations (18) with $N_k^{(0)} = 0$, $\sigma_k^{(0)} = 0$, and with $\mathbf{k}_0^{(0)}$ determined from $\varepsilon_{\mathbf{k}_0} = \omega_p / 2$.

If the matrix elements $T_{\mathbf{k}\mathbf{q}}$ and $S_{\mathbf{k}\mathbf{q}}$ have a definite symmetry, Eqs. (16) and (17) can be solved analytically.

1) The case $T_{\mathbf{k}\mathbf{q}} = T(\mathbf{k} - \mathbf{q})$, $S_{\mathbf{k}\mathbf{q}} = S(\mathbf{k} - \mathbf{q})$. In this case, taking (18) into account, E_k and Δ_k take the form

$$E_k = \varepsilon_k - \varepsilon_{\mathbf{k}_0}, \quad \Delta_k = \hbar(V_k - V_{\mathbf{k}_0}). \quad (19)$$

Substitution of these expressions in (17) yields the magnon

distribution as a function of \mathbf{k}_0 : $[\sigma_{\mathbf{q}}(\mathbf{k}_0)$ and $N_{\mathbf{q}}(\mathbf{k}_0)]$ off the resonance surface. Note that this solution is meaningful (i.e., the quasiparticle spectrum is real) if

$$\hbar < h_i = \min_{\mathbf{q}} |(\varepsilon_{\mathbf{q}} - \varepsilon_{\mathbf{k}_0}) / (V_{\mathbf{q}} - V_{\mathbf{k}_0})|. \quad (20)$$

In fields stronger than h_i an excited magnon system has no stationary state and only vibrational regimes are possible.

On the resonance surface (16) we obtain

$$\varepsilon_{\mathbf{k}_0} - \frac{\omega_p}{2} + 2 \sum_{\mathbf{k} \in \mathbf{K}_0} T(\mathbf{k}_0 - \mathbf{k}) N_{\mathbf{k}} + 2F_N(\mathbf{k}_0) = 0, \quad (21a)$$

$$\hbar V_{\mathbf{k}_0} + \sum_{\mathbf{k} \in \mathbf{K}_0} S(\mathbf{k}_0 - \mathbf{k}) \sigma_{\mathbf{k}} + F_{\sigma}(\mathbf{k}_0) = 0, \quad (21b)$$

where

$$F_N(\mathbf{k}_0) \equiv \sum_{\mathbf{q} \notin \mathbf{K}_0} T(\mathbf{k}_0 - \mathbf{q}) N_{\mathbf{q}}, \quad F_{\sigma}(\mathbf{k}_0) \equiv \sum_{\mathbf{q} \notin \mathbf{K}_0} S(\mathbf{k}_0 - \mathbf{q}) \sigma_{\mathbf{q}}.$$

Simultaneous solution of Eqs. (21) is in general a rather complicated task. These equations, however, can be easily solved if the functions V_k , $S(k)$ and $T(k)$ they contain are independent of the direction of the wave vector \mathbf{k} and if the resonance surface is a sphere of radius k_0 . Then

$$N_{k_0} = [\omega_p / 2 - \varepsilon_{k_0} - 2F_N(k_0)] / 2 \sum_{\mathbf{k} \in \mathbf{K}_0} T(|\mathbf{k}_0 - \mathbf{k}|), \quad (22a)$$

$$\sigma_{k_0} = -[\hbar V_{k_0} + F_{\sigma}(k_0)] / \sum_{\mathbf{k} \in \mathbf{K}_0} S(|\mathbf{k}_0 - \mathbf{k}|). \quad (22b)$$

Substituting these expressions in the condition (14a), we obtain an equation for k_0 . For example, if $V_k = V$, $S(k) = S$, and $T(k) = T$ are constant we have $\Delta_k = 0$ for all k , $\sigma_k = 0$ and $N_k = n_k$ for all $k \neq k_0$, while for $k = k_0$ we obtain

$$\sigma_{k_0} = -\hbar V / \mathcal{M} S, \quad (23a)$$

$$N_{k_0} = \frac{\omega_p / 2 - \varepsilon_{k_0} - 2T \mathcal{N}(\Theta)}{2\mathcal{M} T}, \quad (23b)$$

where

$$\mathcal{M} = \sum_{\mathbf{k} \in \mathbf{K}_0} 1 \approx \frac{1}{\pi} (L k_0)^2$$

is indicative of the "measure" of the resonance surface; L is the linear dimension of the crystal;

$$\mathcal{N}(\Theta) \equiv \sum_{\mathbf{q} \notin \mathbf{K}_0} n_{\mathbf{q}}(\Theta).$$

The value of k_0 is determined from the equation

$$\varepsilon_{k_0} = \frac{\omega_p}{2} - T \left\{ 2\mathcal{N}(\Theta) - \mathcal{M} - 2 \left[\left(\frac{\hbar V}{S} \right)^2 + \mathcal{M}^2 \left(n_{k_0} + \frac{1}{2} \right)^2 \right]^{1/2} \right\}. \quad (24)$$

It must be emphasized that anomalous correlations of magnons (a condensate) are singular only if the coefficients V , S , and T are constant; in all other cases $\sigma_k \neq 0$ for all k . Note also that the equality (23a), rewritten in the form

$$\hbar V + S \sum_{\mathbf{k} \in \mathbf{K}_0} \sigma_{\mathbf{k}} = 0,$$

coincides with the known S -theory result⁸ for "effective" pumping in the limit of infinitesimal dissipation.

2. The case $T_{kq} = t_k t_q$, $S_{kq} = s_k s_q$. We have then

$$E_k = \varepsilon_k - \varepsilon_{k_0} t_k / t_{k_0} - (1 - t_k / t_{k_0}) \omega_p / 2, \quad (25a)$$

$$\Delta_k = h(V_k - V_{k_0} s_k / s_{k_0}). \quad (25b)$$

The equations for N_k and σ_k on the resonance surface are the same as (21), but with $N_q(\mathbf{k}_0)$ and $\sigma_q(\mathbf{k}_0)$ given by expressions (17) and (25) taken into account. Just as above, we get for the spherically symmetric case

$$N_{k_0} = \left(\frac{\omega_p}{2} - \varepsilon_{k_0} - 2t_{k_0} \sum_{q \neq k_0} t_q N_q \right) / 2t_{k_0}^2 \mathcal{M}, \quad (26a)$$

$$\sigma_{k_0} = - \left(hV_{k_0} + s_{k_0} \sum_{q \neq k_0} s_q \sigma_q \right) / s_{k_0}^2 \mathcal{M}, \quad (26b)$$

and the equation for k_0 follows from (14a).

3. EFFECTIVE TEMPERATURE; THERMODYNAMIC STABILITY

The effective temperature Θ reached by a quasiparticle gas after the pump field is switched on can be obtained analytically in two important cases: 1) instantaneous switching and 2) adiabatic switching.

1. When the pump field is switched on instantaneously there is not enough time to change the initial system density matrix ρ_m that describes the equilibrium magnon gas at the temperature Θ_m . This allows us to calculate the energy U_0 of the initially nonequilibrium state of the quasiparticle gas in the rotating coordinate frame, using the functions

$$N_k^{(m)} = \text{Sp}(\rho_m a_k^+ a_k) = [\exp(\varepsilon_k / \Theta_m) - 1]^{-1},$$

$$\sigma_k^{(m)} = \text{Sp}(\rho_m a_k a_{-k}) = 0.$$

Taking the change of the chemical potential into account, we get

$$U_0 = \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} N_{\mathbf{k}}^{(m)}. \quad (27)$$

Since the Hamiltonian (10) does not depend on time explicitly, the energy is conserved. For thermodynamic equilibrium, the expression for the quasiparticle-gas energy is

$$U = \mathcal{E}_0 - \mu \sum_{\mathbf{k}} n_{\mathbf{k}}, \quad (28)$$

where \mathcal{E}_0 is defined by (5) with allowance for relations (12), while the second term is due to the indeterminate number of quasiparticles. The equation

$$U(\Theta) = U(\Theta_m) \quad (29)$$

yields an implicit dependence of Θ and Θ_m . For example, in the case $V_{\mathbf{k}} = V$, $S_{kq} = S$, and $T_{kq} = 0$ this equation takes the form

$$\sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} \left[\exp\left(\frac{\varepsilon_{\mathbf{k}}}{\Theta_m}\right) - 1 \right]^{-1} = - \frac{(hV)^2}{2S}$$

$$+ \sum_{\mathbf{k}} (\varepsilon_{\mathbf{k}} - \varepsilon_0) \left\{ \exp\left[\frac{\varepsilon_{\mathbf{k}} - \varepsilon_0}{\Theta}\right] - 1 \right\}^{-1}. \quad (30)$$

It follows from (30) that the sign of S determines the character of the dependence of the temperature Θ on the amplitude h . For $S > 0$ an increase of h causes Θ to increase, and for $S < 0$ the quasiparticle gas cools down. This cooling continues up to a value h_0 at which Θ reaches zero. With further increase of h it follows from (30) that only a solution with negative Θ is possible. It appears that in this case the parametrically excited system does not reach a stationary state and executes finite motion with $U = U_0 = \text{const}$.

2. If the pump field is turned on adiabatically the entropy is constant. It is assumed that a new quasiparticle-system temperature sets in after each small change of the amplitude h (the chemical potential is determined then from the entropy maximum). The equation for the temperature Θ as a function of Θ_m is

$$\mathcal{P}(\varepsilon_{\mathbf{k}}, \Theta_m) = \mathcal{P}(\Omega_{\mathbf{k}} - \mu, \Theta), \quad (31)$$

where

$$\mathcal{P}(\varepsilon_{\mathbf{k}}, \Theta_m) = \sum_{\mathbf{k}} \left\{ \frac{\varepsilon_{\mathbf{k}}}{\Theta_m} \left[\exp\left(\frac{\varepsilon_{\mathbf{k}}}{\Theta_m}\right) - 1 \right]^{-1} - \ln \left[1 - \exp\left(-\frac{\varepsilon_{\mathbf{k}}}{\Theta_m}\right) \right] \right\}$$

and the expression for $\mathcal{P}(\Omega_{\mathbf{k}} - \mu, \Theta)$ is similar. It is clear that in the general case $\Theta \leq \Theta_m$, but each specific case must be separately computed.

The question of the thermodynamic stability of the equilibrium state of a quasiparticle Bose gas reduces to satisfaction of the following thermodynamic inequalities²⁶: a) the heat capacity at constant volume must be positive, and (b) isotropic compression increases the pressure. An explicit form of condition a) can be obtained by differentiating (28) with respect to Θ :

$$dU/d\Theta > 0.$$

For constant $V_{\mathbf{k}}$ and S_{kq} and for $T_{kq} = 0$ we have, for example,

$$\sum_{\mathbf{k}} (\varepsilon_{\mathbf{k}} - \varepsilon_0)^2 \exp\left[\frac{\varepsilon_{\mathbf{k}} - \varepsilon_0}{\Theta}\right] \left\{ \exp\left[\frac{\varepsilon_{\mathbf{k}} - \varepsilon_0}{\Theta}\right] - 1 \right\}^{-2} > 0.$$

This inequality is obviously always valid.

It is easiest to verify condition b) by using the equation of state of an ideal Bose gas²⁶:

$$P\mathcal{V} = -\Theta \sum_{\mathbf{k}} \ln \left\{ 1 - \exp\left[\frac{\mu - \Omega_{\mathbf{k}}}{\Theta}\right] \right\}.$$

Since the quasiparticle spectrum $\Omega_{\mathbf{k}}$ in our model is independent of the pressure P and of the volume \mathcal{V} , we have $P\mathcal{V} = \text{const}$ for a given temperature. The criterion b) is obviously met in this case.

4. COLLECTIVE OSCILLATIONS

If the quasiparticle gas is thermodynamically stable, small density perturbations relax to an equilibrium within a characteristic time τ determined by the four-particle part of the Hamiltonian (10). The relaxation due to the phase relations is then oscillatory and its frequency ω is referred to as the system collective-oscillation frequency. We work in the

approximation $\omega \gg \tau^{-1}$. We write the time dependences²⁾ of N_k and σ_k in the form

$$i dN_k/dt = \Delta_k \sigma_k^* - \Delta_k^* \sigma_k, \quad (32a)$$

$$i d\sigma_k/dt = 2E_k \sigma_k + (2N_k + 1) \Delta_k, \quad (32b)$$

$$-i d\sigma_k^*/dt = 2E_k \sigma_k^* + (2N_k + 1) \Delta_k^*. \quad (32c)$$

Combining these equations, we readily obtain

$$\frac{d}{dt} \left[|\sigma_k|^2 - \left(N_k + \frac{1}{2} \right)^2 + \left(n_k + \frac{1}{2} \right)^2 \right] = 0. \quad (33)$$

This integral of motion obviously accords with Eq. (14a). In the stationary case it follows from (32a) that σ_k is real, and Eq. (32b) reduces to (14b).

Let us investigate the spectrum of small deviations from equilibrium values of N_k and σ_k :

$$\delta N_k = \delta N_{k\lambda} \exp(\lambda t), \quad \delta \sigma_k = \delta \sigma_{k\lambda} \exp(\lambda t).$$

Linearization of (32a), (32b), and (33) yields

$$i\lambda \delta N_{k\lambda} = \Delta_k (\delta \sigma_{k\lambda}^* - \delta \sigma_{k\lambda}) + \sigma_k (\delta \Delta_{k\lambda} - \delta \Delta_{k\lambda}^*), \quad (34a)$$

$$i\lambda \delta \sigma_{k\lambda} = 2E_k \delta \sigma_{k\lambda} + 2\sigma_k \delta E_{k\lambda} + 2\Delta_k \delta N_{k\lambda} + (2N_k + 1) \delta \Delta_{k\lambda}, \quad (34b)$$

$$\sigma_k (\delta \sigma_{k\lambda} + \delta \sigma_{k\lambda}^*) = (2N_k + 1) \delta N_{k\lambda}, \quad (34c)$$

where

$$\Delta_{k\lambda} = \sum_q S_{kq} \delta \sigma_{q\lambda}, \quad \delta E_{k\lambda} = 2 \sum_q T_{kq} \delta N_{q\lambda}.$$

Equations (34) cannot be solved in the general case. We consider below the case of constant coefficients $V_k = V$, $S_{kq} = S$, and $T_{kq} = T$, when the solution for σ_k is singular. For $k \neq k_0$ we have then from (34)

$$\delta N_{k\lambda} = 0, \quad (35a)$$

$$\delta \sigma_{k\lambda} = (2n_k + 1) S \sum_q \delta \sigma_{q\lambda} / [i\lambda - 2(\varepsilon_k - \varepsilon_{k_0})]. \quad (35b)$$

Summing both sides of (35b) over k we get

$$\sum_k \delta \sigma_{k\lambda} = \mathcal{M} \Lambda \delta \sigma_{k_0\lambda}, \quad \Lambda(\lambda) = \left[1 - S \sum_{k \neq k_0} \frac{2n_k + 1}{i\lambda - 2(\varepsilon_k - \varepsilon_{k_0})} \right]^{-1}. \quad (36)$$

Give this relation, simple transformations of (34) lead to equations for small deviations on the resonance surface:

$$[i\lambda + \mathcal{M} S \Lambda^* (2N_{k_0} + 1)] \delta N_{k_0\lambda} = 2\mathcal{M} S \sigma_{k_0} \operatorname{Re}(\Lambda) \delta \sigma_{k_0\lambda}, \quad (37a)$$

$$[i\lambda - \mathcal{M} S \Lambda (2N_{k_0} + 1)] \delta \sigma_{k_0\lambda} = 4\mathcal{M} T \sigma_{k_0} \delta N_{k_0\lambda}. \quad (37b)$$

From the condition that the Eqs. (37) have nontrivial solutions we obtain an equation for the collective-oscillation spectrum

$$\lambda^2 - 4\mathcal{M} S (N_{k_0} + 1/2) \lambda \operatorname{Im}(\Lambda) + 4[\mathcal{M} S (N_{k_0} + 1/2)]^2 |\Lambda|^2 + 8\mathcal{M}^2 T S \operatorname{Re}(\Lambda) \sigma_{k_0}^2 = 0, \quad (38)$$

whence we get in the single-mode approximation ($\Lambda = 1$) the solution

$$\omega = |\operatorname{Im} \lambda| = 2[(hV)^2 (1 + 2T/S) + (\mathcal{M} S)^2 (n_{k_0} + 1/2)^2]^{1/2}. \quad (39)$$

Equation (38) yields also a stability criterion for the stationary state. Stability is lost in a Hamiltonian system only when λ passes through zero. It follows therefore from (38) that the stationary state is stable if

$$\left(\frac{hV}{\mathcal{M} S} \right)^2 \left[1 + \frac{2T}{S \Lambda(0)} \right] + \left(n_{k_0} + \frac{1}{2} \right)^2 > 0. \quad (40)$$

We conclude this section by noting that Eqs. (32) can be expressed in terms of the Hamiltonian variables N_k and ψ_k :

$$dN_k/dt = -\delta \mathcal{H} / \delta \psi_k, \quad d\psi_k/dt = \delta \mathcal{H} / \delta N_k,$$

where ψ_k is defined by the relation

$$\sigma_k = (N_k^2 + N_k - n_k^2 - n_k)^{1/2} \exp(-i\psi_k),$$

and the Hamiltonian is

$$\begin{aligned} \mathcal{H} = & 2 \sum_k \left(\varepsilon_k - \frac{\omega_p}{2} + \sum_q T_{kq} N_q \right) N_k + 2 \sum_k \left[hV_k \cos \psi_k \right. \\ & \left. + \frac{1}{2} \sum_q S_{kq} (N_q^2 + N_q - n_q^2 - n_q)^{1/2} \cos(\psi_k - \psi_q) \right] \\ & \times (N_k^2 + N_k - n_k^2 - n_k)^{1/2}. \end{aligned} \quad (41)$$

For $n_k = 0$ and $N_k \gg 1$ the expressions for N_k , ψ_k and \mathcal{H} agree with those obtained in the S theory.⁸

DISCUSSION

We have thus shown that the steady-state energy of parametrically excited Bose particles insulated from a thermostat can be represented in a rotating coordinate frame by two terms. The first corresponds to coherent interaction of the magnons with the pump field (condensate), while the second describes an equilibrium quasiparticle gas having a certain effective temperature indicative of the degree of deviation of the magnon distribution function from the coherent one (see Fig. 1). We have obtained criteria for the stability of such a "thermodynamic" state and have found the distribution functions in certain cases.

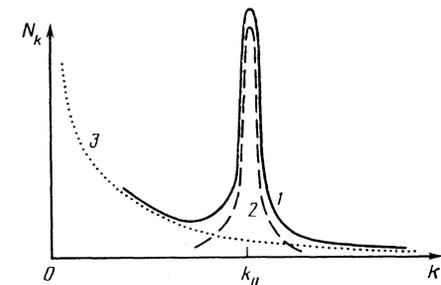


FIG. 1. Nonequilibrium magnon distribution function—curve 1. Curve 2 shows the coherent part of the magnon distribution (condensate), while 3 shows the incoherent part described by the thermodynamic distribution function with effective temperature Θ .

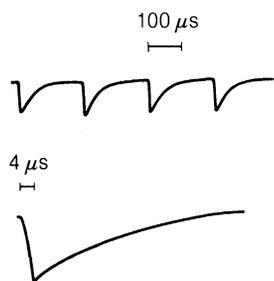


FIG. 2. Example of oscillations of above-threshold susceptibility on a microwave pulse passing through a cavity with a sample of the antiferromagnet CsMnCl_3 (Ref. 28). The lower curve shows the oscillation in enlarged scale. Estimates yield $\tau_m \sim 1 \mu\text{s}$ and $\tau_{\text{ph}} \gtrsim 20 \mu\text{s}$.

The onset of thermodynamic equilibrium in a system of quasiparticles can be revealed in experiment by the saturation effect, when the microwave power absorbed by a sample with pulse-excited magnons decreases noticeable with time. Such behavior of the absorption (see Fig. 2) was observed in a number of studies in which, however, the onset of saturation in the system was accompanied by instability that led to a time-periodic process. According to our theory this means that under the experimental conditions the stationary state was unstable and the system of excited magnons became self-oscillating near this state. Note that saturation is possible also when the quasiparticle gas enters into thermodynamic equilibrium with the heat bath (phonons), if the relativistic interactions are negligibly small.^{11,13} Another criterion of thermodynamic equilibrium in a rotating coordinate frame is quasiparticle accumulation on the band edge of the magnons that can be detected by radiation from the sample.

We note in conclusion that the results can be used to describe real magnetic systems only when the magnons are pumped by a pulsed field having an amplitude much higher than the threshold, so that the energy flow into the heat bath can be neglected. In principle, the thermodynamic approach with a rotating coordinate frame can be generalized (in the context of weak-nonequilibrium thermodynamics) also to include low energy fluxes. We assume that this will lead to a unified description of the below-threshold state, of the threshold, and the of above-threshold state of parametrically excited magnons.

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¹¹In a rotating reference frame the origin of the chemical potential is shifted by $-\omega_p/2$ from its value in the lab.

²²These equations were obtained in the self-consistent-field approximation by a method similar to that used in Ref. 20. Analogous equations were analyzed earlier in Ref. 11.

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