Phase transition leading to the appearance of spontaneous coherence in a system of interacting oscillators and fields

G. M. Zaslavskiĭ, Yu. A. Kudenko, and A. P. Slivinskiĭ

Physics Institute, Siberian Division, USSR Academy of Sciences
(Submitted January 15, 1975)
Zh. Eksp. Teor. Fiz. **68**, 2276–2289 (June 1975)

For a system with resonance interaction between oscillators and the radiation field, the conditions that lead to a phase transition to a state of spontaneous coherence below T_c are obtained. Thermodynamic averages and the correlation function of the oscillators are calculated. An analogous phase transition is detected in systems with a process of the stimulated Raman-scattering type, in which a phase transition in the field also exists, and in a system of interacting waves of the decay type.

PACS numbers: 03.70.

1. INTRODUCTION

In a system of atoms interacting with the radiation field, at sufficiently long wavelengths cooperative effects begin to play an important role. That state of the atoms-field system in which collective properties of the atoms are manifested in spontaneous radiative processes was first studied by Dicke[1] in a model of two-level atoms and was called the superradiative state. Recently, an investigation of the thermodynamic properties of the Dicke model, carried out by Hepp and Lieb[2], led to an important result: the transition of the system from the normal to the superradiative state is a phase transition analogous to ferromagnetic ordering (cf. also[3]). For temperatures $T < T_c$ and sufficiently strong interaction of the atoms with the field there appears a macroscopic number of field photons (proportional to the number N of atoms), and the mean number of photons per atom, y_0, which is the order parameter, becomes nonzero. An analogous property is also possessed by three-level systems[4,5], in which a new effect arises: a succession of phase transitions at temperatures T_1 and T_2, each of which is associated with the opening of the corresponding radiation channel[4] (the "cascade" character of the appearance of a macroscopic number of photons).

The results described above point to a new possibility for creating systems possessing coherent properties. In this connection, it is of interest to study a wide class of physical systems, and the present paper is devoted to this. It will be shown below that resonant processes of interaction of the field with atoms and with other fields can lead under certain conditions to a second-order phase transition, manifested in the appearance of spontaneous coherence of the oscillations of the atoms when $T < T_c$ (analogous to the spontaneous magnetization in a system of spins). In the paper we consider the following classical systems that are of interest for applications: a system of oscillators interacting with the radiation field (Sec. 2), a system of the stimulated Raman scattering (SRS) type (Sec. 3) and a system of waves with interaction of the decay type (Sec. 4); the conditions under which the phase transition to the coherent state occurs are obtained. The comparative simplicity of the systems investigated (compared with the purely quantum Dicke model) has made it possible to elucidate in detail the characteristic features of the phase transition, to calculate the correlation function, etc. At the end of Sec. 2 and of the whole article, certain possibilities of observing the phase transition described are discussed.

2. SUPERRADIATIVE PHASE TRANSITION IN A SYSTEM OF OSCILLATORS

We shall write the Hamiltonian describing a system of classical oscillators interacting with a field in the form

$$H=\omega_k\alpha_k\alpha_k^*+\omega_0\sum_j^N J_j+\frac{g}{\sqrt{N}}\sum_j^N(V_{kj}\alpha_k^*c_j+V_{kj}^*\alpha_kc_j^*), \qquad (2.1)$$

where α_k is the complex amplitude of the field having frequency ω_k; $J_j = |c_j|^2$ is the action of the j-th oscillator; $V_{kj} = e^{ikr_j}$, k is the wave number and r_j is the coordinate of the j-th oscillator. The interaction in (2.1) is of the dipolar type, and we consider the case close to resonance ($\omega_k \approx \omega_0$). The interaction constant $g = \sqrt{\rho\omega_k}$, where μ is the dipole moment of an individual oscillator and ρ is the volume density of the oscillators.

It is not difficult to calculate the partition function of the system (2.1):

$$Z=\int_{-\infty}^{\infty}d\alpha\,d\alpha^*\prod_{j=1}^N\int_0^\infty dJ_j\int_0^{2\pi}d\theta_j\exp(-\beta H), \qquad (2.2)$$

where $c_j = J_j^{1/2}e^{i\theta_j}$ and $\beta = 1/T$. Putting $\alpha = |\alpha|e^{i\varphi}$ and $\psi_j = \theta_j - \varphi$, we rewrite (2.2):

$$Z=2\pi\int_0^\infty d|\alpha|^2\exp(-\beta\omega_k|\alpha|^2)\prod_{j=1}^N\int_0^\infty dJ_j\int_0^{2\pi}d\psi_j$$
$$\times\exp\left[-\beta\omega_0J_j-2\beta\frac{g}{\sqrt{N}}|\alpha|J_j^{1/2}\cos(\psi_j+kr_j)\right]$$
$$=2\pi N\int_0^\infty dy\exp(-\beta\omega_kNy)\left\{2\int_0^\infty dJe^{-\beta\omega_0J}I_0(2\beta g\sqrt{yJ})\right\}^N$$
$$=\left(\frac{2}{\beta\omega_0}\right)^N\frac{2\pi}{\beta\omega_k(1-g^2/\omega_k\omega_0)} \qquad (2.3)$$

where $y = |\alpha|^2/N$ and $I_0(x)$ is the Bessel function of imaginary argument. As can be seen from (2.3), there is no phase transition in the classical limit under consideration.

As will be seen below, a phase transition turns out to be possible if for the oscillators there exists a classical analog of an "energy gap." In this case the integration over J in (2.3) is performed between the limits $J = J_0$ and ∞, where J_0 is found from the condition $H \geq V_0$. A classical system with an energy gap is nonequilibrium and can be represented easily by means of the following model. Let the trajectories of an oscillator in the phase plane have a stable limit cycle C (see Fig. 1). From a physical point of view this means that in the energy region II (Fig. 1), on the average, an ordinary frictional force acts on the oscillators, while in the region I, on the average, negative friction acts. If we now assume that all the dissipative parameters are sufficiently small compared with the time of the interaction of the oscillators with the field, we can study the thermodynamics of an excited system of this type. In this case, thermal fluctuations can carry oscillators over from region II to region I and vice versa, overcoming the effective potential barrier between them. The effect of the occupation of the low-energy region by particles can be neglected if the barrier height is sufficiently large compared with T. The latter condition has the form

$$\beta\omega_0 J_0 \approx \beta V_0 \gg 1. \tag{2.4}$$

Thus, slow nonequilibrium processes will slightly smear out the phase transition considered below.

Another case in which an energy cutoff appears ($H \geq V_0$) may be associated with nonlinearity of the oscillators[1)].

With the assumptions made, we have, approximately, in place of (2.3),

$$Z \approx 2\pi N \int_0^\infty dy\, e^{-\beta\omega_k y N} \left\{ 2 \int_{J_0}^\infty dJ\, e^{-\beta\omega_0 J} I_0(2\beta g \sqrt{yJ}) \right\}^N$$

$$= 2\pi N \int_0^\infty dy \exp\{N[-\beta\omega_k y + \ln Q(y)]\},$$

$$Q(y) = 2 \int_{J_0}^\infty dJ\, e^{-\beta\omega_0 J} I_0(2\beta g \sqrt{yJ}). \tag{2.5}$$

For $N \to \infty$ the value of Z is determined by the extremum:

$$Z \xrightarrow[N\to\infty]{} \text{const}\cdot\exp\{N[-\beta\omega_k y_0 + \ln Q(y_0)]\}, \tag{2.6}$$

where y_0 satisfies the equation

$$\beta\omega_k = \frac{1}{Q(y_0)} \frac{dQ(y_0)}{dy}. \tag{2.7}$$

If (2.7) has no solution, then

$$Z \to \text{const}\cdot\exp\{N[-\beta\omega_k y + \ln Q(y)]\}_{y=0},$$

which, according to (2.5), indicates the absence of a contribution to the partition function from the interaction. The quantity y_0, on the other hand, is equal to

$$y_0 = N^{-1}\langle|\alpha|^2\rangle$$

and has the meaning of the mean number of photons per oscillator. Thus, if there exists a T_c such that for $T < T_c$ the quantity $y_0 \neq 0$, while, for $T > T_c$, $y_0 = 0$, then a phase transition occurs to the superradiative state (for $T < T_c$) of the oscillators, analogous to that which occurs in two-level systems[2,3].

We shall consider Eq. (2.7), using the definition (2.5):

$$\frac{\omega_k}{g}\sqrt{y_0} = \frac{1}{Q(y_0)} \int_{J_0}^\infty dJ \sqrt{J}\, e^{-\beta\omega_0 J} I_1(2\beta g \sqrt{y_0 J}). \tag{2.8}$$

Equation (2.8) is the analog of the Curie-Weiss equation, and we shall show that for $\beta > \beta_c$ ($T < T_c$) it has a nontrivial solution $y_0 \neq 0$. Expanding $I_1(x)$ in the region of $y_0 = 0$, we have, for the determination of T_c, the equation

$$\omega_0\omega_k/g^2 = 1 + \beta_c\omega_0 J_0. \tag{2.9}$$

From (2.9), in particular, it can be seen that for $J_0 = 0$ there is no nontrivial solution. Rewriting (2.9) in the form

$$T_c = \omega_0 J_0 (\omega_0\omega_k/g^2 - 1)^{-1} = V_0(\omega_0\omega_k/g^2 - 1)^{-1}, \tag{2.10}$$

we find that a phase transition exists only under the condition that the interaction is sufficiently weak:

$$g^2 < \omega_0\omega_k. \tag{2.11}$$

This condition is the opposite of that which was found in[2,3] for a system of two-level atoms interacting with the radiation field.

We shall now make use of the inequality (2.4). From (2.8) follows the equation

$$\frac{\omega_k}{g}\sqrt{\frac{y_0}{J_0}} = \frac{I_1(2\beta g \sqrt{y_0 J_0})}{I_0(2\beta g \sqrt{y_0 J_0})}, \tag{2.12}$$

determining y_0 for $T < T_c$, and the expression (2.10) for T_c goes over into

$$T_c = J_0 g^2/\omega_k = V_0 g^2/\omega_0\omega_k. \tag{2.13}$$

The graphical solution of Eq. (2.12) for y_0 is given in Fig. 2, and

$$y_0(T=0) = J_0 g^2/\omega_k^2. \tag{2.14}$$

We shall now find $\langle J \rangle$, by making use of the expressions (2.5), (2.6):

$$\langle J\rangle = -\frac{1}{\beta}\lim_{N\to\infty}\frac{1}{N}\frac{\partial \ln Z}{\partial\omega_0} = -\frac{1}{\beta}\frac{\partial}{\partial\omega_0}\ln Q(y_0,\omega_0)$$

$$= \int_{J_0}^\infty J dJ e^{-\beta\omega_0 J} I_0(2\beta g\sqrt{y_0 J}) \left[\int_{J_0}^\infty dJ e^{-\beta\omega_0 J} I_0(2\beta g \sqrt{y_0 J})\right]^{-1}. \tag{2.15}$$

The dependence of $\langle J \rangle$ on T that follows from formula (2.15) is shown in Fig. 3, in which the Rayleigh-Jeans distribution for $g = 0$ is drawn with the dashed line, and

$$\langle J\rangle|_{T=0} = \langle J\rangle|_{T=T_c} = \frac{V_0}{\omega_0}\left(1 + \frac{g^2}{\omega_0\omega_k}\right). \tag{2.16}$$

As can be seen from (2.16) and Fig. 3, for $T < T_c$ the number of excited oscillators exceeds the equilibrium value for $g = 0$. This is connected with the capture of photons, the density of which for $T < T_c$ is equal to $y_0 \neq 0$ (Fig. 2).

It is not difficult to understand the meaning of formula (2.13) if we notice that the potential energy of the dipolar interaction of the field with an oscillator is of the order of $U \sim \mu\sqrt{J_0}\,\mathscr{E}$, where the intensity of the

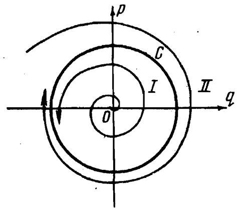

FIG. 1

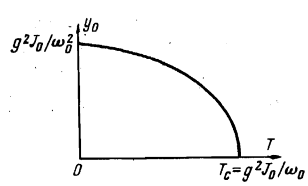

FIG. 2

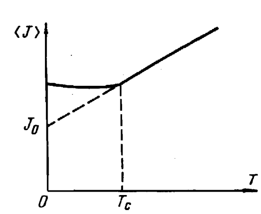

FIG. 3

electric field of the oscillators $\mathscr{E} = \rho\mu\sqrt{J_0}$. Taking into account that $g = \mu\sqrt{\rho\omega_k}$, we obtain $U \sim \rho\mu^2 J_0 = g^2 J_0/\omega_k = T_c$. Thus, for $T > U = T_c$ thermal fluctuations destroy the bound state of the oscillators with the field.

We can also analyze the physical meaning of the phase transition by turning our attention to the interaction term

$$H_{int} \sim \sum_j \sqrt{J_j}\cos(\psi_j + kr_j). \quad (2.17)$$

Although, under the resonance condition, the phase ψ_j is an invariant of the motion for each site separately, ($\dot{\psi}_j$ is an invariant of the motion for each site separately, ($\dot{\psi}_j = \omega_k - \omega_0 \neq 0$), it can take random values that differ greatly from site to site and the averaging (2.17) at sufficiently high T gives $(\langle H_{int}^2\rangle)^{1/2} \sim N^{1/2}$. On the other hand, for low T the formation of a bound state of the oscillators with the field is favorable. In this case the variation of the phase of the oscillators at the different sites becomes coherent and acquires spatial periodicity, with period equal to the wavelength of the photons. Then $(\langle H_{int}^2\rangle)^{1/2} \sim N$.

We shall calculate the spatial correlator of the displacements q and velocities $\dot{q} = p$ of the oscillators:

$$R_m(q) = \lim_{N\to\infty} \frac{1}{N}\sum_{l=1}^{N}\langle q_l q_{l+m}\rangle, \quad R_m(p) = \lim_{N\to\infty}\frac{1}{N}\sum_{l=1}^{N}\langle p_l p_{l+m}\rangle.$$

Making use of the relations (the mass of an oscillator equals 1)

$$q = \sqrt{1/2\omega_0}\,(c^* + c), \quad p = i\sqrt{\omega_0/2}\,(c^* - c),$$

we write

$$R_m(q) = \lim_{N\to\infty}\frac{1}{NZ\omega_0}\sum_{l=1}^{N}\langle J_l^{1/2} J_{l+m}^{1/2}\cos(\theta_l - \theta_{l+m})\rangle$$

$$= \lim_{N\to\infty}\frac{1}{NZ\omega_0}\sum_{l=1}^{N}\langle J_l^{1/2} J_{l+m}^{1/2}\cos(\psi_l - \psi_{l+m} - kma)\rangle, \quad (2.18)$$

where $a = r_{l+1} - r_l$ is the spacing between the oscillators of the "lattice." We write out the integration in (2.18) analogously to (2.3):

$$R_m(q) = \frac{1}{\omega_0}\lim_{N\to\infty}\frac{1}{NZ_N}\sum_{l=1}^{N}\cos kma\int_0^\infty d(Ny)\exp[-\beta\omega_k Ny + (N-2)\ln Q_l(y) + 2\ln\bar{Q}_l(y)], \quad (2.19)$$

where $Q_l(y)$ is the same as in (2.5), and

$$\bar{Q}_l(y) = \int_{J_0}^{\infty}\sqrt{J_l}\,dJ_l\int_0^{2\pi}d\psi_l\exp[-\beta\omega_l J_l - 2\beta g\sqrt{y_0 J_l}\cos\psi_l]$$

$$= 2\int_{J_0}^{\infty}dJ_l J_l^{1/2} e^{-\beta\omega_0 J_l} I_1(2\beta g\sqrt{yJ_l}). \quad (2.20)$$

Transformations in (2.19) give

$$R_m(q) = \frac{1}{\omega_0}\cos kma\cdot\lim_{N\to\infty}\frac{1}{Z_{N-2}}\int_0^\infty d(Ny)\exp\left\{N[-\beta\omega_k y + \ln Q(y)] + 2\ln\left[\frac{\bar{Q}(y)}{Q(y)}\right]\right\} = \frac{1}{\omega_0}\left[\frac{\bar{Q}(y_0)}{Q(y_0)}\right]^2\cos kma, \quad (2.21)$$

where y_0 is again determined by Eq. (2.8), since the equation for the saddle point for $N \to \infty$ goes over into (2.8). If we make use of the identity

$$\frac{dQ(y)}{dy} = \frac{\beta g}{\sqrt{y}}\bar{Q}(y),$$

then, taking (2.8) into account, we obtain from (2.21), finally,

$$R_m(q) = \frac{\omega_k^2}{\omega_0 g^2}y_0(T)\cos kma, \quad (2.22)$$

where $y_0(T)$ is the solution of Eq. (2.8). As can be seen from (2.22), the correlator is proportional to the order parameter $y_0(T)$, and vanishes for $T > T_c$. It oscillates as a function of distance, with the dimensionless period $2\pi/ka = \lambda/a$. The appearance of a periodic structure with the photon wavelength λ is obvious, inasmuch as emission and absorption are the only source of the interaction between the oscillators.

In analogy with (2.22), we can obtain

$$R_m(p) = \frac{\omega_0\omega_k^2}{g^2}y_0(T)\sin kma. \quad (2.23)$$

If we make use of the inequality (2.4), the expression (2.22) and (2.23) can be written in a more explicit form:

$$R_m(q) = \frac{1}{\omega_0}\frac{I_1^2(2\beta g\sqrt{y_0 J_0})}{I_0^2(2\beta g\sqrt{y_0 J_0})}\cos kma,$$

$$R_m(p) = \omega_0\frac{I_1^2(2\beta g\sqrt{y_0 J_0})}{I_0^2(2\beta g\sqrt{y_0 J_0})}\sin kma. \quad (2.24)$$

In conclusion we shall give some qualitative estimates to compare the conditions for a phase transition in a system of two-level atoms with those in a system of oscillators. If we introduce the critical density of atoms, $\rho_c = \hbar\omega/\mu^2$, in the two-level case the threshold condition for superradiation has the form[2,3] $\rho > \rho_c$, and, e.g., for the optical region ($\omega \sim 10^{15}$ Hz, $\mu \sim 10^{-18}$ esu) we have $\rho_c \sim 10^{24}\ \text{cm}^{-3}$; ρ_c increases with increase of ω.

The case we have considered, of a system of classical oscillators, may turn out to be more favorable in the region of optical and higher frequencies, since, according to (2.11), the phase transition occurs under the opposite condition $\rho < \rho_c$. It follows from (2.13) that

$$T_c/T_0 = \rho/\rho_c, \quad (2.25)$$

where $T_0 = \hbar\omega J_0$. Since, according to (2.4), $T_c/T_0 \ll 1$, we can evidently obtain a superradiative state at sufficiently low densities ρ of oscillators. For example, in the optical region ($\rho_c \sim 10^{24}\ \text{cm}^{-3}$) and with $T_0 \sim 10^4$ K, for the densities of impurities in a solid ($\rho \sim 10^{21}\ \text{cm}^{-3}$) we obtain $T_c \sim 10$ K. We then have the following range for the density ρ:

$$\rho_c \geq \rho \geq T_c/J_0\mu^2.$$

3. THE PHASE TRANSITION IN STIMULATED RAMAN SCATTERING

As the simplest generalization of the phase transition to the superradiative state we shall consider SRS of an external field by oscillators c_j. We write the Hamiltonian of the system in the form

$$H = \omega\alpha\alpha^* + \omega_0\sum_j c_j c_j^* + \omega_1 b_1 b_1^* + \omega_2 b_2 b_2^* + \frac{g_1}{\sqrt{N}}\sum_j V_{1j}c_j\alpha b_1^* + \frac{g_2}{\sqrt{N}}\sum_j V_{2j}c_j^*\alpha b_2^* + \text{c.c.}, \quad (3.1)$$

where α is the amplitude of the external field, with frequency ω, being scattered; b_1 and b_2 are the amplitudes of the field in the anti-Stokes and Stokes scattering respectively (see Fig. 4); $g_{1,2}$ are the constants of the corresponding interaction processes (the moduli of the matrix elements of the processes, the phase factors of which are written as V_{1j}, V_{2j}); $\omega_1 = \omega + \omega_0$; $\omega_2 = \omega - \omega_0$. At sufficiently high field intensities b_1, b_2, emission and absorption processes have comparable probabilities, and it is this which leads to the necessity of treating the thermodynamic system of oscillators and resonance fields b_1, b_2 as a single whole. Moreover, the external field α and its frequency ω are not too large, so that a thermodynamic formulation of the problem has meaning.

We rewrite (3.1) in the form

$$H=N\omega y+\omega_0\sum_j J_j+\omega_1|b_1|^2+\omega_2|b_2|^2+2g_1\sqrt{y}\sum_j\sqrt{J_j}\,|b_1|\cos(\theta_{V_1}+\theta_{c_j}+\theta_\alpha-\theta_1)+2g_2\sqrt{y}\sum_j\sqrt{J_j}\,|b_2|\cos(\theta_{V_2}-\theta_{c_j}+\theta_\alpha-\theta_2), \quad (3.2)$$

where y and J are the same as in Sec. 2 and the θ are the phases of the corresponding components. We shall make use of the following transformation of volumes:

$$\begin{aligned} d\theta_\alpha d\theta_{c_j}d\theta_1 d\theta_2 &= 2d\theta_1 d\theta_2 d(\theta_\alpha-\theta_{c_j})\,d(\theta_\alpha+\theta_{c_j}) \\ &= 8d(\theta_\alpha+\theta_{c_j}+\theta_1)\,d\psi_1' d(\theta_\alpha-\theta_{c_j}+\theta_2)\,d\psi_2' \\ &= 8d(\theta_\alpha+\theta_{c_j}+\theta_1)\,d(\theta_\alpha-\theta_{c_j}+\theta_2)\,d\psi_1 d\psi_2, \end{aligned}$$
$$\psi_1'=\theta_\alpha+\theta_{c_j}-\theta_1,\quad \psi_2'=\theta_\alpha-\theta_{c_j}-\theta_2,\quad \psi_{1,2}=\psi_{1,2}'+\theta_{V_{1,2}}. \quad (3.3)$$

Now, in analogy with (2.2), (2.3) and (2.5), we can write an expression for the partition function:

$$\begin{aligned} Z &= (32\pi^2)^N N^2 e^{-\beta\omega N y}\int_0^\infty dy_1\int_0^\infty dy_2\exp[-\beta N(y_1\omega_1+y_2\omega_2)] \\ &\cdot\left\{\int_{J_0}^\infty dJ\,e^{-\beta\omega_0 J}\int_0^{2\pi}d\psi_1\int_0^{2\pi}d\psi_2\exp[-2\beta\sqrt{yJ}\,(g_1\sqrt{y_1}\cos\psi_1+g_2\sqrt{y_2}\cos\psi_2)]\right\}^N \\ &= (32\pi^2)^N N^2 e^{-\beta\omega N y}\int_0^\infty dy_1\int_0^\infty dy_2\exp[-\beta N(\omega_1 y_1+\omega_2 y_2)+N\ln Q(y_1,y_2)], \end{aligned}$$
$$Q(y_1,y_2)=4\int_{J_0}^\infty dJ\,e^{-\beta\omega_0 J}I_0(2\beta g_1\sqrt{yy_1J})\,I_0(2\beta g_2\sqrt{yy_2J}),\qquad y_{1,2}=|b_{1,2}|^2/N. \quad (3.4)$$

In the limit $N\to\infty$ we have

$$Z\sim\text{const}\cdot\exp[-\beta N(\omega_1 y_{01}+\omega_2 y_{02})+N\ln Q(y_{01},y_{02})], \quad (3.5)$$

where the saddle points y_{01}, y_{02} satisfy the system

$$\beta\omega_1 Q(y_{01},y_{02})=\frac{\partial Q(y_{01},y_{02})}{\partial y_1},\quad \beta\omega_2 Q(y_{01},y_{02})=\frac{\partial Q(y_{01},y_{02})}{\partial y_2}. \quad (3.6)$$

If the system (3.6) has no solution, the following three cases are then possible: $y_{01}=0$ and the equation

$$\beta\omega_2 Q(0,y_{02})=\frac{\partial Q(0,y_{02})}{\partial y_2} \quad (3.7)$$

has a solution with $y_{02}\neq 0$, or $y_{02}=0$ and the equation

$$\beta\omega_1 Q(y_{01},0)=\frac{\partial Q(y_{01},0)}{\partial y_1} \quad (3.8)$$

has a solution with $y_{01}\neq 0$, or none of Eqs. (3.6)—(3.8) has a solution, and then $y_{01}=y_{02}=0$ in (3.5).

As in the preceding section, we can convince ourselves that for $J_0\neq 0$ there are no nontrivial solutions at sufficiently high temperatures. Thus, as T is lowered a phase transition can arise to a state with an anomalously large photon scattering density. These states may be called superscattering states. They were introduced and investigated in[7], but this is, apparently, the first time that a temperature phase transition to a superscattering state has been noted.

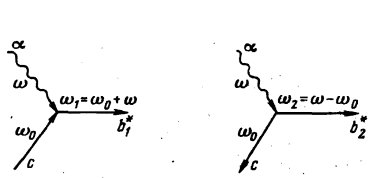

FIG. 4

Investigation of the system (3.6) in the general case is cumbersome, although, on the basis of the results of[4], we can point out the following qualitative behavior of the solutions of (3.6). With decrease of T scattering channels at frequencies ω_1, ω_2 are successively opened at temperatures T_1, T_2 respectively. The relationship between T_1 and T_2 depends on the constants g_1, g_2. If, e.g., $T_2 > T_1$ (which is realized for $g_2 > g_1$), then, for $T > T_2$, $y_{01}=y_{02}=0$ and there is no superscattering; for $T_1 < T < T_2$, $y_{01}=0$, $y_{02}\neq 0$ and is determined from (3.7), and the superscattering is of the Stokes type; for $T < T_1$, the quantities y_{01}, $y_{02}\neq 0$ and are found from (3.6). In the latter case superscattering occurs with respect to both components of the field.

In the limiting case of large values of J_0, when, according to (2.4), $\beta\omega_0 J_0 \gg 1$ and $\beta g_{1,2}\sqrt{yy_{1,2}}J_0 \ll \beta\omega_0 J$, the expression for $Q(y_1, y_2)$ is simplified:

$$Q(y_1,y_2)=Q(y_1)Q(y_2),\qquad Q(y_i)=I_0(2\beta g_i\sqrt{yy_iJ_0})\quad(i=1,2). \quad (3.9)$$

The system (3.6) decouples into two independent equations:

$$\frac{\omega_i\sqrt{y_{0i}}}{g_i\sqrt{yJ_0}}=\frac{I_1(2\beta_i g_i\sqrt{yy_{0i}J_0})}{I_0(2\beta_i g_i\sqrt{yy_{0i}J_0})}. \quad (3.10)$$

From these equations we can determine the two critical temperatures $T_{1,2}$:

$$T_i=J_0 g_i^2 y/\omega_i\quad(i=1,2). \quad (3.11)$$

It follows from (3.11) that the ratio of the critical temperatures for Stokes (T_2) and anti-Stokes superscattering (T_1) is equal to

$$\frac{T_2}{T_1}=\left(\frac{g_2}{g_1}\right)^2\frac{\omega_1}{\omega_2}=\left(\frac{g_2}{g_1}\right)^2\frac{\omega+\omega_0}{\omega-\omega_0}. \quad (3.12)$$

The principal difference between the expressions (3.11) and (2.13) is connected with the fact that the phase-transition temperature in the SRS is proportional to the intensity y of the pumping field and is thereby accessible to external control. The expressions (y_{01}, y_{02} for $T=0$) for the maximum density of photons of the coherent fields are modified analogously:

$$y_{0i}=\frac{g_i^2}{\omega^2}yJ_0, \quad (3.13)$$

i.e., the intensity of the coherent field is proportional to the pumping-field intensity.

If the temperature is fixed it follows from (3.10) that there exists a phase transition with respect to the pumping-field intensity parameter. A coherent state of the oscillator arises for fields

$$y>y_c=\frac{\omega_i T}{g_i^2 J_0}. \quad (3.14)$$

4. SYSTEMS OF THE DECAY TYPE (THREE-WAVE AND FOUR-WAVE PROCESSES)

The results obtained above enable us to go over to one of the most interesting and important cases—a system in which processes of resonant decay and coalescence of waves occur. We write the Hamiltonian of the system in the form

$$H=\sum_k[\omega_k^{(1)}J_k^{(1)}+\omega_k^{(2)}J_k^{(2)}]+\omega_0 J^{(0)}+\sum_k[V_k a_0 b_k^{(1)}b_{k+k_0}^{(2)*}+\text{c.c.}],$$
$$b_k^{(1)}=\sqrt{J_k^{(1)}}\exp(i\theta_k^{(1)}),\quad b_k^{(2)}=\sqrt{J_k^{(2)}}\exp(i\theta_k^{(2)}),\quad a_0=\sqrt{J^{(0)}}\exp(i\theta_0), \quad (4.1)$$

$$\dot{\theta}_k^{(1)}=\omega_k^{(1)},\quad \dot{\theta}_k^{(2)}=\omega_k^{(2)},\quad \dot{\theta}_0=\omega_0,$$

where a_0 is the complex amplitude of a monochromatic field with wave-number k_0, and the resonance conditions

$$\omega_{k+k_0}^{(2)}=\omega_k^{(1)}+\omega_0. \tag{4.2}$$

are assumed to be fulfilled.

The indices (1), (2) refer respectively to the two branches of oscillations between which a photon with frequency ω_0 is transferred. We shall also assume that

$$\beta\omega^{(1)}J_0^{(1)}\gg\beta\omega^{(2)}J_0^{(2)}\gg 1, \tag{4.3}$$

where $J_0^{(1),(2)}$ are the minimum values of the action of the waves. The first inequality in (4.3), as is well-known[8], leads to the possibility of using the quasi-harmonic approximation for the waves. The second inequality in (4.3), as will be clear below, is a necessary condition for the existence of a phase transition. Inasmuch as the waves with frequencies $\omega^{(2)}$ and ω_0 in the process (4.2) are completely exchanged by the actions under the condition (4.3)[8], the existence of a lower band on the action of the wave ($\bar{y} = J_0^{(0)}/N = J_0^{(2)}$) also follows from the second inequality in (4.3). Of interest, however, is the case $\beta\omega_0\bar{y} \ll 1$, for which it is possible to obtain values of T_c that are not too small. On the other hand, the existence of the lower bound $\bar{y}$ for the order parameter y means that, if it exists, the phase transition to the coherent state is a first-order phase transition, close to being a second-order phase transition.

The appearance of the wave-action cutoff parameters $\bar{y}$, $J_0^{(1)\,(2)}$ is connected, as in the case of oscillators, with the existence of metastable excited states of the oscillations. It is simplest to imagine such states in a plasma or in another unstable medium. The development of instability of any mode leads to an increase in its amplitude, up to a certain stationary (metastable) value, and the weak interaction of such modes creates small temperature fluctuations[9]. Inasmuch as the increments in the instability depend on k, the lower bounds $J_{0,k}^{(1)}$ and $J_{0,k_0+k}^{(2)}$ are also functions of k.

We turn to the calculation of the partition function for the Hamiltonian

$$H=\omega_0 J^{(0)}+\sum_k\left[\omega_k^{(1)}J_k^{(1)}+\omega_{k_0+k}^{(2)}J_{k_0+k}^{(2)}\right]+\frac{2}{N^{1/2}}\sum_k G\sqrt{J^{(0)}J_k^{(1)}J_{k_0+k}^{(2)}}\cos\psi_k,$$

$$V_k=\frac{G_k}{\sqrt{N}}e^{i\varphi},\quad \psi_k=\varphi+\theta_0+\theta_k^{(1)}-\theta_{k_0+k}^{(2)}, \tag{4.4}$$

where N is the number of modes over which the summation is performed; in addition, a finite number of terms, making no contribution to the interaction, has been omitted in the unperturbed part of H. We have

$$Z=N\int_{\bar{y}}^{\infty}dy\int_0^{2\pi}d\varphi\prod_k\int_{J_{0,k}^{(1)}}^{\infty}dJ_k^{(1)}\int_{J_{0,k_0+k}^{(2)}}^{\infty}dJ_{k_0+k}^{(2)}\int_0^{2\pi}d\theta_k^{(1)}\int_0^{2\pi}d\theta_{k_0+k}^{(2)}\exp(-\beta H),\quad y=J^{(0)}/N, \tag{4.5}$$

where the limits of integration are chosen in accordance with the analysis performed above.

We shall make use of the following transformation formula for the elements of integration over the angles:

$$d\varphi\, d\theta_k^{(1)}d\theta_{k_0+k}^{(2)}=2d\varphi\, d(\theta_k^{(1)}+\theta_{k_0+k}^{(2)})\,d(\theta_k^{(1)}-\theta_{k_0+k}^{(2)})=2d(\theta_k^{(1)}+\theta_{k_0+k}^{(2)})\,d\varphi\, d\psi_k. \tag{4.6}$$

Substituting (4.6) into (4.5) and taking into account that H depends only on ψ_k, we find

$$Z=N(8\pi^2)^N\int_{\bar{y}}^{\infty}dy\,e^{-\beta\omega_0 Ny}\prod_k Q_k(y),$$

$$Q_k(y)=\int_{J_{0,k}^{(1)}}^{\infty}dJ_k^{(1)}\int_{J_{0,k_0+k}^{(2)}}^{\infty}dJ_{k_0+k}^{(2)}\int_0^{2\pi}d\psi_k\exp\{-\beta[\omega_k^{(1)}J_k^{(1)}+\omega_{k_0+k}^{(2)}J_{k_0+k}^{(2)}$$
$$+2G_k\sqrt{yJ_k^{(1)}J_{k_0+k}^{(2)}}\cos\psi_k]\}=2\int_{J_{0,k}^{(1)}}^{\infty}dJ_k^{(1)}\int_{J_{0,k_0+k}^{(2)}}^{\infty}dJ_{k_0+k}^{(2)}\cdot$$
$$\cdot\exp\{-\beta[\omega_k^{(1)}J_k^{(1)}+\omega_{k_0+k}^{(2)}J_{k_0+k}^{(2)}]\}\,I_0(2\beta G_k\sqrt{yJ_k^{(1)}J_{k_0+k}^{(2)}}). \tag{4.7}$$

Finally, we have

$$Z\sim\text{const}\cdot\exp\left\{-N\left[\beta\omega_0 y_0-\frac{1}{N}\sum_k\ln Q_k(y_0)\right]\right\}, \tag{4.8}$$

and the extremum point y_0 is found from the equation

$$\beta\omega_0=\lim_{N\to\infty}\frac{1}{N}\sum_k\frac{d}{dy}[\ln Q_k(y_0)]=\int d\omega\,\rho(\omega)\frac{d}{dy}[\ln Q_\omega(y_0)], \tag{4.9}$$

where $\rho(\omega)$ is the density of states (frequencies) per unit frequency interval and unit volume, and Q_ω is obtained from Q_k by means of the change of variables. For $T > T_c$ Eq. (4.9) has no solution $y_0 \geq \bar{y}$, and in (4.8) we must put $y_0 = 0$.

The critical temperature T_c can be obtained from (4.9) by expanding $Q_k(y_0)$ in the vicinity of $y_0 = \bar{y}$, taking into account the inequality $\beta_c\bar{y}\omega_0 \ll 1$:

$$\beta_c\omega_0=\lim_{N\to\infty}\frac{1}{N}\sum_k\frac{G_k^2}{\omega_k^{(1)}\omega_{k_0+k}^{(2)}}(1+\beta_c\omega_k^{(1)}J_{0,k}^{(1)})(1+\beta_c\omega_{k_0+k}^{(2)}J_{k_0+k}^{(2)})$$
$$=\int d\omega\rho(\omega)\frac{G_\omega^2}{\omega(\omega+\omega_0)}[1+\beta_c\omega J_{0,\omega}][1+\beta_c(\omega_0+\omega)J_{0,\omega_0+\omega}]$$

or, taking the inequalities (4.3) into account:

$$T_c=\frac{1}{\omega_0}\int d\omega\rho(\omega)G_\omega^2 J_{0,\omega}^{(1)}J_{0,\omega_0+\omega}^{(2)}=G^2J_0^{(1)}J_0^{(2)}/\omega_0, \tag{4.10}$$

where G and J_0 are certain averages (over the wave packet) of the quantities G_ω and $J_{0,\omega}$. Inasmuch as the dimensionless quantity $J_0 \gg 1$ by virtue of the classical nature of the problem, the value of T_c from (4.10) is greater than in the case of oscillators interacting with a field with the same value of the interaction constant (in (2.13)).

As in the case of oscillators interacting with a field, below T_c "capture" (binding) of one of the types of wave occurs ($y_0 \neq 0$). Quanta with frequency ω_0 are emitted and absorbed by two branches ($J_k^{(1)}$ and $J_k^{(2)}$), and this leads, below T_c, to an anomalously large value of the average three-wave interaction energy. To determine the equilibrium value of y_0 we substitute (4.7) into (4.9) and take into account the inequalities (4.3):

$$\omega_0\sqrt{y_0}=\int d\omega\rho(\omega)G_\omega\sqrt{J_{0,\omega}^{(1)}J_{0,\omega_0+\omega}^{(2)}}\,\frac{I_1\left(2\beta G_\omega\sqrt{y_0J_{0,\omega}^{(1)}J_{0,\omega_0+\omega}^{(2)}}\right)}{I_0[2\beta G_\omega(y_0J_{0,\omega}^{(1)}J_{0,\omega_0+\omega}^{(2)})^{1/2}]}, \tag{4.11}$$

in which $y_0 \geq \bar{y}$. In the case when G and J_0 do not depend on ω (on k), Eq. (4.11) is analogous to (2.12). This makes it possible to use for estimates the expressions (2.14)–(2.16), in which we must replace $g \to G\sqrt{J_0}$.

We remark also that the process of establishing coherence of the phases of oscillating modes with different values of k below T_c is accompanied by the appearance of coherent structures in coordinate space.

The results obtained for three-wave processes have a universal character. It is not difficult, e.g., to generalize them to the case of four-wave interaction in conditions of an external pumping field c_q at frequency

Ω. In the presence of parametric amplification of the field with frequency ω_0, for H the interaction term in the Hamiltonian (4.4) must be replaced by

$$H_{int}=\sum_{k_1,k_2} V_k a_0 b_{k_1}^{(1)} b_{k_2}^{(2)*} c_q^* + \text{c.c.},$$

$$k_0+k_1\approx k_2+q, \quad \omega_0+\omega_{k_1}^{(1)}\approx\omega_{k_2}^{(2)}+\Omega. \qquad (4.12)$$

On parametric conversion of the frequencies we have, in place of (4.12),

$$H_{int}=\sum_{k_1,k_2} V_k a_0 b_{k_1}^{(1)} b_{k_2}^{(2)*} c_q + \text{c.c.},$$

$$k_0+k_1+q\approx k_2, \quad \omega_0+\omega_{k_1}^{(1)}+\Omega\approx\omega_{k_2}^{(2)}. \qquad (4.13)$$

In both cases (4.12) and (4.13), the phase transition consists in the appearance of a nonzero average field intensity y_0 at the frequency ω_0 and in a sharp increase, below T_c, in the average cross-sections of the corresponding processes—of the "superamplification" and "superconversion" types. The quantitative results have the appearance of the expressions (3.13), (3.14) in which G must be replaced by $G|c_q|$.

5. CONCLUSION

The investigation carried out above shows that, generally speaking, under certain conditions a threshold value with the character of a phase transition may be associated with an arbitrary resonance process between a system of the thermodynamic type (i.e., having an effective thermal scatter of the parameters) and a certain nonthermodynamic field. This transition occurs with respect to the temperature or the external field and is manifested in the appearance of spontaneous coherent properties in the system below T_c. Another interpretation of the transition is that, for $T > T_c$, the density of the intensity (i.e., of the number of photons) of the "scattering field" which mediates the resonant interaction between elements of the thermodynamic part of the system is of the order of $N^{-1/2}$, whereas below T_c the intensity density remains finite as $N\to\infty$. The thermodynamic properties of the system must be understood in a fairly general sense, since the system can be strongly excited on the average. In this case the effective temperature of the system characterizes the degree of fluctuational scatter of the parameters and may not coincide with the real temperature of the medium (such a situation is realized, e.g., in SRS).

We have considered the most typical cases of resonant systems. We must discuss in more detail systems in which SRS occurs. Strictly speaking, the real situation differs somewhat from that considered, since usually the nonlinearity is very strong and the excitation parameter J_0, unlike the one considered, is a function of the pumping amplitude. However, the general picture, leading to a threshold value as a result of the competition between the ordering resonant interaction between the atoms and the field and the disordering action of the thermal fluctuations, should be conserved. In view of this, it is useful to make certain estimates. We shall write, e.g., the equation of motion for the Stokes component of the field[8] in the symmetrized form:

$$\dot{B}_S=in\left(R\frac{\partial\alpha}{\partial R}\right)\frac{(\omega_S\omega_v\omega_L)^{1/2}}{A_{0v}}B_L B_v^*,$$

$$B_S=A_S/\sqrt{\omega_S}, \quad B_L=A_L/\sqrt{\omega_L}; \quad B_v=A_v/\sqrt{\omega_v},$$

where A_S, A_L, and A_v are the amplitudes of the scattively. A_{0v} is the level of excitation of the field of the molecules, R is the generalized coordinate of a molecule, α is the susceptibility of the molecules and n is the density of molecules. Then, by means of (3.1) and (3.14), it is not difficult to obtain the following expression for the critical pumping field at which the phase transition occurs:

$$A_{Lc}^2=\mathscr{E}_c=T_{\text{eff}}\Big/\left[n\left(R\frac{\partial\alpha}{\partial R}\right)^2\right],$$

where $\mathscr{E}_c$ is the pumping-energy density. For the typical values $A_L^2\sim10^6$–10^7 esu, $n\sim10^{22}$ cm^{-3}, $R\,\partial\alpha/\partial R\sim10^{-24}$ esu, we obtain: $T_{\text{eff}} < 10^{-19}$ esu $= 10^{-3}$ deg. The actual determination of the quantity T_{eff} presents certain difficulties, exactly as does the relation of T_{eff} with the emission linewidth, but the estimate obtained for T_{eff} is only two orders of magnitude smaller than the energy corresponding to the linewidth of the molecular vibrations. If we take into account that the linewidth depends essentially on the pumping amplitude and narrows sharply at large A_L, the estimate obtained for T_{eff} may turn out to be completely realistic.

In conclusion it must be said that the results obtained demonstrate the theoretical possibility of using thermodynamic ("noisy") systems to obtain coherent fields, the variation in the intensity of which is close in character to a second-order phase transition. This transition should also be accompanied by growth of fluctuations near the transition point and by an increase of the relaxation time, which may be detected, e.g., by means of photon echo.

The authors consider it their pleasant duty to thank I. M. Lifshitz for his interest and for discussions in the process of carrying out the work, and S. A. Akhmanov, V. A. Ignatchenko and Yu. L. Klimontovich for useful discussions and critical comments.

[1]An investigation of the conditions for a phase transition in this case was carried out for ferroelectrics in [6].

[1] R. H. Dicke, Phys. Rev. 93, 99 (1954).
[2] K. Hepp and E. H. Lieb, Ann. Phys. 76, 360 (1973).
[3] Y. K. Wang and F. Hioe, Phys. Rev. A7, 831 (1973).
[4] G. M. Zaslavskiĭ, Yu. A. Kudenko, and A. P. Slivinskiĭ, Preprint IFSO-17F (Physics Institute, Siberian Division), 1974; Fiz. Tverd. Tela 16, 3391 (1974) [Sov. Phys.-Solid State 16, 2197 (1975)].
[5] T. M. Makhviladze, A. I. Rez, and M. E. Sarychev, FIAN (Physics Institute of the Academy of Sciences) Preprint No. 15, 1974.
[6] K. S. Aleksandrov, I. P. Aleksandrova, G. M. Zaslavskiĭ, A. V. Sorokin, and V. F. Shabanov, ZhETF Pis. Red. 21, 58 (1975) [JETP Lett. 21, 27 (1975)].
[7] U. Kh. Kopvillem, Proc. 2nd Conference on Solid State Physics, Ashkhabad, 1969; O. N. Gadomskiĭ, and V. R. Nagibarov, Zh. Eksp. Teor. Fiz. 62, 896 (1972) [Sov. Phys.-JETP 35, 475 (1972)]; T. M. Makhviladze and L. A. Shelepin, Izv. AN SSR 37, 2190 (1973) [Bull. Acad. Sci. USSR 37, (1973)].
[8] N. Bloembergen, Nonlinear Optics, Benjamin, N. Y., 1965 (Russ. transl. "Mir", M., 1966).
[9] B. B. Kadomtsev, in the Collection "Voprosy teorii plazmy (Problems in Plasma Theory)", Vol. 4, Atomizdat, M., 1964; A. A. Galeev and N. Z. Sagdeev, in the Collection "Voprosy teorii plazmy (Problems in Plasma Theory)", Vol. 7, Atomizdat, M., 1973.

Translated by P. J. Shepherd
242