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NONEQUILIBRIUM PHASE TRANSITION IN LASER-ACTIVE MEDIA*

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The master equation for a single-mode laser is solved exactly in a relevant limit. It is shown that a nonequilibrium phase transition of the laser *atoms* occurs in the limit of a large number of atoms. The phase transition analogies in laser light are reinterpreted as the consequence of this transition.

STRIKING analogies to second-order phase transitions have been found in the properties of laser light near the threshold.[1–3] This is of particular interest because the laser is an open system far away from thermal equilibrium.

In the present paper the cooperative nature of the laser transition is discussed. Within a single-mode laser model it is shown that a non-equilibrium phase transition of the laser *atoms* occurs in the limit of a large number of atoms. The phase transition analogies in laser light are reinterpreted as the consequence of this transition which is similar to the transition of the Curie–Weiss model[4] for a ferromagnet. In particular the concept of the thermodynamic limit (number of atoms $\to \infty$) is shown to play the same role as in equilibrium phase transitions. This result is obtained from an exact stationary solution of the laser master equation in the limit of small reservoir coupling constants.

We deal with a system of N two-level atoms and a single-mode radiation field with the total Hamiltonian $H = H_{\text{Atoms}} + H_{\text{Field}} + H_I$. In the dipole and rotating wave approximation[5] the interaction Hamiltonian H_I, in terms of creation and annihilition operators, is given by

$$H_I = i\hbar\mu \sum_{m=1}^{N} [b^+(a_1^+a_2)_m - b(a_2^+a_1)_m] \quad (1)$$

where a running wave mode is assumed.[6] μ is proportional to the atomic dipole moment. In Lax's stochastic laser model[7] the interaction with the surroundings (pump, lossy resonator mirrors etc.) is taken into account by coupling heat reservoirs to the atoms and the field. We use this model and start from the basic master equation for the density operator ρ in the form used by Gordon[8]

$$\dot{\rho} = -\frac{i}{\hbar}[H,\rho] + \sum_{ij} w_{ij} A^{ij}(\rho) + \gamma F(\rho). \quad (2)$$

In (2) we have abbreviated the atomic and field reservoir terms by A^{ij} and F. For $i \neq j$, w_{ij} are the probabilities for reservoir-induced transitions from level j to i. γ is the field decay constant. $F(\rho)$ contains the thermal parameter $\bar{n} = [\exp(\hbar\omega/kT) - 1]^{-1}$. For more details see references 7 and 8.

By means of a perturbation calculation with respect to the reservoir terms we have found[9] the exact stationary solution of (2) in the limit $(w_{21}, w_{12}, \gamma) \to 0$. The resulting diagonal elements of the density matrix in the energy representation are

$$p(n,m) = p(0,0)\binom{N}{m}\prod_{k=1}^{n+m} \frac{w_{21} + \gamma\bar{n}f_N(k)}{w_{12} + \gamma(\bar{n}+1)f_N(k)}, \quad (3)$$

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$$f_N(k) = \sum_{\nu=0}^{k} (k-\nu)\binom{N}{\nu} \Big/ \sum_{\nu=0}^{k} \nu \binom{N}{\nu}. \qquad (4)$$

$p(n,m)$ denotes the probability to find n photons in the light field and m atoms in the upper level. $p(0,0)$ is determined by normalization. We emphasize that the limit $(w_{21}, w_{12}, \gamma) \to 0$ is basically different from the case $w_{21} = w_{12} = \gamma = 0$. In the latter case the stationary solution of (2) is not unique. This ambiguity is removed by a weak coupling of the atom-field system to the external reservoirs – similar as in perturbation theory for degenerate systems. Here we deal with a system between two heat baths of different temperatures; even arbitrary small coupling constants make the system tend to a unique stationary state which is determined essentially by the *ratios* of the coupling constants. Note that the effect due to the finite values of w_{ij} and γ is very small for real lasers.[10]

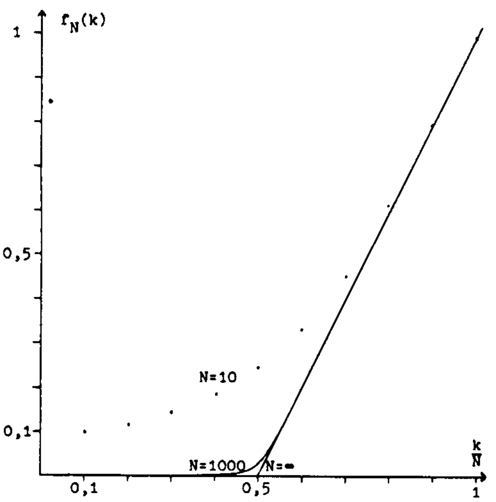

FIG. 1. $f_N(k)$ as a function of k/N for various N. For $k \geq N$, $f_N(k) \equiv (2k-N)/N$ for all N.

Our result (3) which is exact for all N, now permits a reliable analysis of the role of the atoms in the laser transition. The crucial quantity is the function $f_N(k)$ (Fig. 1). It shows a nontrivial N-dependence which is specific for the laser. The region around $k = N/2$ where f_N has a mathematical singularity for $N \to \infty$, turns out to be directly related to the threshold, the critical point of the laser phase transition. The threshold may be defined by that value of w_{21} ($\sim$ pump power) at which the photon distribution $p(n) = \sum_m p(n,m)$ begins to develop a maximum for $n > 0$. From (3) we find the N-dependent threshold condition $w_{21} \simeq w_{12} + \gamma f_N(N/2)$ where $f_N(N/2) \sim N^{-1/2}$. On the other hand the leading terms of the mean occupation numbers $\langle m_1 \rangle$ and $\langle m_2 \rangle$ as calculated from (3)

$$\frac{\langle m_2 \rangle}{\langle m_1 \rangle} = \begin{cases} w_{21}/w_{12} - 0\{N^{-1}\} & (w_{21} < w_{12}) \\ 1 \quad - 0\{e^{-N}\} & (w_{21} > w_{12}) \end{cases} \qquad (5)$$

suggest the threshold condition $w_{21} = w_{12}$. Thus in the threshold definition there exists an uncertainty $w_{12} \leq w_{21} \leq w_{12} + \gamma f_N(N/2)$ which, although not of practical relevance, gives the key for a reinterpretation of the phase transition analogies in laser light. Evidently, the sharpening to a theoretically unique threshold for $N \to \infty$ corresponds to the fact in equilibrium phase transitions that only in the thermodynamic limit there exists a well defined critical temperature T_c. This many-particle effect is reflected in the pump power dependence of the atomic occupation numbers at threshold (Fig. 2). The crucial role of the number of atoms is equally well substantiated by the N-dependence of the mean photon number for $N \to \infty$ (Fig. 2)[11]

$$\langle n \rangle \longrightarrow \begin{cases} w_{21}/(w_{12} - w_{21}) & (w_{21} < w_{12}) \\ N(w_{21} - w_{12})/2\gamma & (w_{21} > w_{12}). \end{cases} \qquad (6)$$

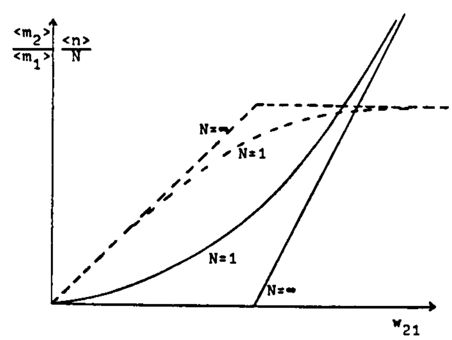

FIG. 2. The mean relative photon number (solid lines) and the ratio of the mean occupation numbers (dotted lines) as functions of w_{21} ($\sim$ pump power) for $N \to \infty$ and $N = 1$ (qualitatively).

These results clearly demonstrate the analogy of the number of laser atoms to the number of particles in equilibrium phase transitions, and they suggest that we regard the laser atoms – not the laser light – as the primary system from the viewpoint of a cooperative phenomenon. Therefore we introduce an order parameter in terms of the atomic variables. Indeed, because the polarization $\vec{P}$ of the laser-active medium is directly coupled to the light amplitude, there is no macroscopic contribution to $\vec{P}$ below threshold according to (6), but above threshold a macroscopic polarization $\vec{P}(\vec{r}, t) = \vec{p} \exp i(\vec{k}\vec{r} - \omega t)$ occurs. Thus above a critical pump power, well defined for $N \to \infty$, the effective interaction between the atomic dipole moments (by way of the laser light) leads to an ordered nonequilibrium state of the system of atoms characterized by the order parameter $\vec{p}$, the coherent polarization of the laser-active medium. Assuming $|\vec{p}|$ to be proportional to the field amplitude $\sim (\langle n \rangle / V)^{1/2}$, one obtains from (6) $\vec{p} = 0$ below and $|\vec{p}| \sim (w_{21} - w_{12})^{1/2}$ above threshold in the 'thermodynamic' limit $N, V \to \infty$ (with constant N/V),[12] i.e. a critical exponent $\beta = 1/2$. This molecular-field like behaviour confirms the result of the approximate Fokker–Planck method[5] which leads to a Landau theory for the transition in the light field.[2,3] Here the laser light plays the role of the field coupled to the order parameter, quite analogous e.g. to the static electric field which is caused by the spontaneous polarization of ferroelectrics below T_c. Accordingly the variable conjugate to $\vec{P}$ is an external polarized light field of definite phase and frequency ω (for example that injected by another laser) which breaks the symmetry of the phase of the dipole moments.

The single-mode laser model used in this paper automatically leads to a long-range atomic interaction which is independent of the dimensionality of the system. Indeed, the Hamiltonian (1) leads to an effective interaction which couples all atoms equally to one another. In this respect, (1) is similar to the interaction energy of the Curie–Weiss model for a ferromagnet[4] which displays a phase transition of the molecular-field type. Contrary to this artificial model, a single-mode laser model is fairly realistic.

Similar as in equilibrium theory, singularities occur in the 'thermodynamic' limit introduced above. Some of them are shown in Figs. 1 and 2. On a macroscopic intensity scale, laser output measurements indeed exhibit a discontinuity of the first order derivative of the light intensity at threshold.[2,13]

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5. For a review of laser theory see e.g. HAKEN H., *Handbuch der Physik* XXV/2c, Springer, Berlin, Heidelberg, New York (1970).
6. The spatial modulation $\sim \exp(i\vec{k}\vec{r}_m)$ has been eliminated, cp. reference 5 p. 114, 117.
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9. A detailed derivation of this solution will be published elsewhere.
10. This effect is essentially a shift of the threshold parameters. The relative change is of the order of magnitude $(\Sigma w_{ij} + \gamma)\gamma/4\mu^2 N$, i.e. $10^{-6} - 10^{-10}$ (reference 5 p. 107). Note that $\mu^2 N \sim N/V$.

11. This asymptotic N-dependence is also obtained from the approximate Fokker–Planck solution, reference 5 p. 159.

12. An appropriate *discrete* sequence of N, V ensures the existence of the mode frequency ω.

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Die Mastergleichung für einen einmodigen Laser wird in einem relevanten Grenzfall exakt gelöst. Es wird gezeigt, daß ein Nichtgleichgewichtsphasenübergang der Laser*atome* im Grenzfall vieler Atome auftritt. Die Phasenübergangsanalogien im Laserlicht werden als Folge dieses Überganges interpretiert.