First- and second-order phase transitions in the Dicke model: Relation to optical bistability

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The authors consider a system comprised of a collection of identical two-level atoms interacting with the electromagnetic field in the dipole approximation and with an externally applied steady-state coherent driving field. The atoms are considered to interact with each other only via the electromagnetic field and are assumed to be contained within a volume much smaller than a resonance wavelength. The system is treated as a quantum-statistical ensemble in the rotating frame, rotating at the carrier frequency of the externally applied coherent field. The Hamiltonian in this frame is explicitly time independent and the exact analog of a spin temperature is defined. The thermodynamic-Green's-function method is used to determine the thermodynamic equilibrium properties of the system in the rotating frame. This results in a nonlinear relation between the applied field and the field in the volume containing the atoms. The expression is a nonlinear function of the effective or "spin" temperature as well, and has the form of a first-order phase transition with the macroscopic atomic polarization as order parameter. In the low-"spin"-temperature limit, and for perfect tuning, the expression reduces to the exact form of the nonlinear relation for optical bistability derived by Bonifacio and Lugiato and relating the incident and transmitted fields for atoms in a ring cavity in the mean-field approximation. The results contain absorptive as well as dispersive contributions. These results predict essentially different behavior as a function of off tuning compared with behavior predicted by others using statistical steady-state models far from thermodynamic equilibrium. In the limit of zero value for the applied field amplitude, the results reduce to the conditions for the well-known second-order "superradiant" phase transition. The condition for the existence of the second-order phase transition is shown to depend on the cavity tuning and the photon escape rate. The authors also show why, in light of the experiments in optical bistability, this second-order phase is difficult to observe.

I. INTRODUCTION

The use of a saturable absorber to induce optical bistability (hereafter referred to as OB) was suggested independently by a number of persons in articles published some years ago.¹⁻⁴ Recently, the phenomenon was demonstrated and studied experimentally by Gibbs, McCall, and Venkatesan⁵ who used a cw dye laser to excite atoms of sodium vapor in a cell between the plates of a Fabry Perot interferometer. They observed a nonlinear dependency of the transmitted field as a function of the dye-laser input which exhibited hysteresis, differential gain, and bistable behavior dependent upon certain experimental and material conditions.

Aside from the interest in OB from the practical applications aspect as the optical analog of the transistor, optical clipper or limiter, and optical memory element, the recent demonstration of the phenomenon⁵ has generated considerable interest from the fundamental standpoint as a rather clear example of spontaneous ordering in an open, stationary system of matter interacting with light. This has led to much recent theoretical activity.

The first model for OB was suggested by McCall,⁶ who introduced a nonlinear susceptibility

and used the Maxwell-Bloch representation in the full propagation treatment. His results are necessarily calculational rather than analytical. Bonifacio and Lugiato⁷ (hereafter referred to as BL), were the first to obtain the main features of OB from implementation of the Maxwell-Bloch model, which they solved analytically in closed form. They have since extended their model⁸ to a quantum-statistical representation of a stationary system far from thermodynamic equilibrium, but with the same essential assumptions, the most crucial being the "mean-field" approximation, which amounts to requiring that the field be sufficiently uniform over the volume of active atoms. From this model, BL obtain additional manifestations of the phenomenon, such as line shape, spectral distribution, and critical slowdown in the region of bistable behavior. More recently, the limits of validity of the mean-field approximation and the assumptions connected with the boundary conditions associated with the cavity have been systematically examined by Meystre,⁹ who integrated the six coupled Maxwell-Bloch equations to compare the forward and backward wave amplitudes as a function of position between the Fabry Perot mirrors.

Of particular note here is the work of Walls

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et al.,¹⁰ who in several papers discuss the main features of OB which are on the whole in agreement with the results of BL. They also consider as a model a statistical stationary system far from thermodynamic equilibrium, but the main approximation in their model is that the atoms of the active material are contained within a volume much smaller than a resonance wavelength of the atomic transition. In this way, they remove the explicit requirements for the boundary condition imposed by a cavity. Their mean-field condition is therefore quite different in principle from that of BL. In this case the mean-field approximation means a factorization of the moments for products of atomic-shift operators involving different atoms. This is precisely what we mean by the "mean-field" approximation in the model we present in this paper.

The observation of OB and the results of the theoretical activity cited above are strongly suggestive of an interpretation of the phenomenon as a first-order phase transition in the lightmatter interaction. The quantum-theoretical models cited previously^{8,10} can be looked upon as extensions of Dicke's earlier model¹¹ for superradiance. It is well known that Dicke's model Hamiltonian (having no externally applied field) leads to a second-order phase transition in thermodynamic equilibrium. The instability in the ground state of the Dicke Hamiltonian and the condition for the existence of the second-order phase transition in thermodynamic equilibrium were first discussed by Hepp and Lieb,¹² who obtained exact, closed-form results. Others later introduced more tractable calculational methods for studying the equilibrium thermodynamic properties of the Dicke model.¹³⁻¹⁵ Since then, many authors have analyzed the equilibrium thermodynamics of various modifications of the Dicke model.¹⁶ In particular, Gilmore and Bowden¹⁵ discussed the effect of an externally applied field on the second-order phase transition and the circumstances under which such a field can induce a first-order phase transition.

It would appear that the phenomenon of optical bistability as a first-order phase transition and the thermodynamic equilibrium properties of the Dicke Hamiltonian should in some way be related. The first attempt to relate the equilibrium and nonequilibrium properties of the Dicke model was performed by Gilmore and Narducci.¹⁷ The equilibrium conditions do not lead to bistability in the presence of an externally applied field in their model calculations.

Most of the theoretical effort thus far has been directed to the description of absorptive bistability, yet the results of the experiments of Gibbs et al.⁵ indicate a drastic asymmetry in the output for off tuning of the cavity frequency from the fixed incident dye-laser frequency, with the dyelaser frequency slightly detuned from the atomic transition. This suggests the strong dominance of dispersive contributions. Recently, BL^{18} and Hassan et al.¹⁹ have examined fully quantum mechanically the effects of dispersive contributions to OB for both homogeneously and inhomogeneously broadened systems in a ring-cavity configuration far from thermodynamic equilibrium.

Because of the observed strong contribution of dispersive as opposed to absorptive OB, the results of the experiments suggest that a system exhibiting OB need not necessarily be far from thermodynamic equilibrium. Also, it appears reasonable to us that the existence of the secondorder "super-radiant" phase transition in thermodynamic equilibrium and the existence of OB as a first-order phase transition should stem from the same basic matter-light interaction, and would therefore be expected to arise as separate limiting cases of a single model.

If these suppositions are correct, then it is expected that the first- and second-order phase transitions should result from the nonzero and zero values for the externally applied field, respectively, in the same basic model. However, in order to discuss thermodynamic equilibrium properties in the presence of a time-dependent external field, we must introduce a "spin" temperature. Furthermore, we would expect the results of such a model for OB to reduce to the results of the nonequilibrium models in the limit of zero effective temperature. Owing to these considerations, we have made a fresh examination of the thermodynamic properties of the Dicke model.

In Sec. II, the Hamiltonian for a collection of two-level atoms in the small-volume limit. coupled to an applied field in a coherent state, is presented and the effective interaction which gives rise to collective atomic behavior in a cavity is discussed. Section III is used to present the thermodynamic Green's functions and associated correlation functions pertinent for the determination of collective behavior in the system. In Sec. III we derive the conditions for a firstorder phase transition, which is the main result of this work, and discuss the conditions of OB behavior in terms of system parameters and effective, or "spin" temperature. The connection between the first- and second-order phase transitions in the limit of zero applied field is discussed in Sec. IV, where we show that the second-order phase transition in Refs. 13-16 is actually a special case of the OB discussed

II. HAMILTONIAN AND EFFECTIVE ATOMIC INTERACTION

We consider a collection of N identical twolevel atoms with energy levels $\epsilon_2 > \epsilon_1$ interacting with the electromagnetic field in the dipole approximation. The electromagnetic field which interacts with the atoms is comprised of an internal field and, in addition, an applied cw field assumed to be in a coherent state,²⁰ with field amplitude α and carrier frequency ω_0 . The atoms of the material are considered to be confined to a volume V_g which is much smaller than the resonance wavelength λ_r associated with the atomic barestate separation. This assumption does not critically affect our results and tends to allow emphasis upon the nature of collective atomic aspects in the results in an unencumbered fashion.

The Hamiltonian *H* is written in the form

$$H = H_0 + H', \qquad (2.1)$$

where, in units such that

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$$n = c = 1,$$

$$H_{0} = \sum_{k=0}^{\infty} \omega_{k} a_{k}^{\dagger} a_{k} + \sum_{\substack{n=1,2\\j=1}}^{N} \epsilon_{n} C_{nj}^{\dagger} C_{nj},$$
(2.2a)

and

$$H' = \sum_{j=1}^{N} \sum_{k=0}^{\infty} g_k a_k^{\dagger} C_{1j}^{\dagger} C_{2j} + \text{H.c.}$$
$$+ \alpha^* e^{i\omega_0 t} \sum_{j=1}^{N} C_{1j}^{\dagger} C_{2j} + \text{H.c.} \qquad (2.2b)$$

The first and second terms in (2.2a) represent the free-field and the free-atomic system, respectively, whereas the first two terms in (2.2b) describe the interaction of the atoms with the internal field and the last two terms describe their interaction with the external field. The field creation and annihilation operators conform to Bose commutation relations,

$$[a_k, a_l^{\dagger}] = \delta_{kl}, \quad [a_k, a_l] = 0,$$
 (2.3a)

and the atomic operators are Bose operators 21 which satisfy

$$[C_{jl}, C_{ik}^{\dagger}] = \delta_{ji} \delta_{kl}, \quad [C_{jl}, C_{ik}] = 0.$$
 (2.3b)

In order to discuss the thermodynamic equilibrium properties of (2.2) it is necessary to eliminate the explicit time dependency appearing in (2.2b). To accomplish this we canonically transform to the frame rotating at rate ω_0 and discuss the thermodynamics of (2.2) in the rotating frame. In this frame the applied electromagnetic field appears as a dc field. The unitary transformation U which we desire is explicitly

$$U = \exp\left(\frac{1}{2}i\omega_{0}t\sum_{j=1}^{N} (C_{2j}^{\dagger}C_{2j} - C_{1j}^{\dagger}C_{1j}) + i\omega_{0}t\sum_{k=0}^{\infty} a_{k}^{\dagger}a_{k}\right), \qquad (2.4)$$

which gives

$$H_T = UHU^{-1} \tag{2.5a}$$

after canonical frequency renormalization. If we write (2.5a) as

$$H_{T} = H_{0T} + H_{T}', \qquad (2.5b)$$

then, explicitly

$$H_{0T} = \sum_{k=0}^{\infty} \Omega_k a_k^{\dagger} a_k + \frac{1}{2} \sum_{\substack{n=1,2\\j=1}}^{N} \Omega_n C_{nj}^{\dagger} C_{nj}, \qquad (2.6a)$$

$$H'_{T} = \sum_{j=1}^{N} \sum_{k=0}^{\infty} g_{k} a_{k}^{\dagger} C_{1j}^{\dagger} C_{2j} + \text{H.c.} + \alpha^{*} \sum_{j=1}^{N} C_{1j}^{\dagger} C_{2j} + \text{H.c.},$$
(2.6b)

where

$$\Omega_k = \omega_k - \omega_0, \quad \Omega_2 = \epsilon - \omega_0, \quad \Omega_1 = \omega_0 - \epsilon$$
 (2.7)

and

$$\epsilon = \epsilon_2 - \epsilon_1, \quad \epsilon_2 > \epsilon_1.$$
 (2.8)

We shall apply the thermodynamic Green'sfunction method in the next section to analyze the thermodynamic equilibrium properties of (2.6). To this end it is necessary to adiabatically eliminate the field variables from the interaction Hamiltonian (2.6b). The Heisenberg equations of motion for the field variables must therefore be integrated. From (2.6) we obtain for the equations of motion in the rotating frame

$$i\frac{\partial a_{k}^{\dagger}}{\partial t} + \tilde{\Omega}_{k}a_{k}^{\dagger} = -g_{k}^{*}\sum_{j=1}^{N}C_{2j}^{\dagger}C_{1j}, \qquad (2.9)$$

where

$$\tilde{\Omega}_{b} = \Omega_{b} + i\gamma_{b}$$

and the phenomenological damping γ_k (rate of photon escape from the quantization volume V) has been introduced to account for the reflectivity of cavity mirrors which define the volume of quantization. A formal integral of (2.9) yields

$$a_{k}^{\dagger}(t) = ig_{k} \sum_{j=1}^{N} \int_{-\infty}^{t} dt' C_{2j}^{\dagger}(t') C_{1j}(t') \times e^{i\tilde{\Omega}_{k}(t-t')}.$$
(2.10)

The equation of motion for C_{2j}^{\dagger} yields

$$C_{2i}^{\dagger}(t) = C_{2i}^{\dagger}(t') e^{i/2\overline{\Omega}_{2}(t-t')}, \quad t > t', \quad (2.11)$$

where we have defined

$$\overline{\Omega}_{\frac{1}{2}} = \pm \left(\Omega_{\frac{1}{2}}^{2} + |\Delta|^{2}\right)^{1/2}, \qquad (2.12)$$

the renormalized energy due to the interaction of the atoms with the total field. It is crucial at this point to include the average Stark contribution $|\Delta|^2$, to be self-consistently determined later. The assumption (2.11) is consistent with the requirement that the system be Markovian for times τ , $\tau = t - t'$, long compared with some characteristic relaxation time τ_{c^*} . In this case, explicitly,

$$\tau_{c_k} = \gamma_k^{-1} \, .$$

Contributions to the integral (2.10) are therefore expected to be dominated by values of the integral corresponding to times t' in the neighborhood of the upper limit. Thus, if (2.11) is used in (2.10)and the indicated integration is performed, we obtain

$$a_{k}^{\dagger}(t) = -\frac{g_{k}^{*}}{\lambda_{k}} \sum_{j} C_{2j}^{\dagger}(t) C_{1j}(t), \qquad (2.13)$$

where

$$\lambda_{k} = \Omega_{k} + i\gamma_{k} - \frac{1}{2}(\overline{\Omega}_{2} - \overline{\Omega}_{1}). \qquad (2.14)$$

If (2.13) is used in (2.6b), the result is

$$H'_{T} = -\sum_{k=0}^{\infty} \sum_{\substack{j,l \ j \neq l}} |g_{k}|^{2} \left(\frac{1}{\lambda_{k}} + \frac{1}{\lambda_{k}^{*}}\right) C_{2l}^{\dagger} C_{1l} C_{1j}^{\dagger} C_{2j}$$
$$+ \left(\alpha^{*} \sum_{j} C_{1j}^{\dagger} C_{2j} + \text{H.c.}\right), \quad (2.15)$$

where it is understood that the self-energy terms, i.e., those for which j = l, are now included in the diagonal part of the Hamiltonian, H_{0T} . These terms make only a small contribution, i.e., O(1/N). The first term in the above expression is the analog of spin-spin interaction in the Heisenberg model for ferromagnetism. It is also formally identical with the interaction among Cooper pairs in the Bardeen-Cooper-Schrieffer theory of superconductivity.^{22,23} This specific form for the interaction was shown earlier to be responsible for the collective atomic behavior which results in the second-order super-radiant phase transition in thermodynamic equilibrium,²⁴ and is formally identical to the retarded dipoledipole interaction which has been shown to be responsible for the collective and individual atom radiation reaction and radiative frequency shifts in the dynamic process of superfluorescent relaxation.25,26

At this point, it is expedient to take the sum over modes k in (2.15) to an integral, i.e.,

$$\sum_{k} \to V \int \frac{d^{3}k}{(2\pi)^{3}} = \frac{V}{(2\pi)^{3}} \int k^{2} dk d\Omega, \qquad (2.16)$$

where V is the quantization volume and $d\Omega$ is the element of solid angle at the center of symmetry of the volume V. The sum over modes in (2.15) thus becomes an integral I, i.e.,

$$I = \frac{V}{(2\pi)^3} \int_{4\pi} d\Omega \int_0^\infty d\omega |g(\omega)|^2 \\ \times \left(\frac{1}{\omega - \omega_0 - \sigma + i\gamma} + \frac{1}{\omega - \omega_0 - \sigma - i\gamma}\right),$$
(2.17)

where

$$\sigma \equiv \frac{1}{2} (\overline{\Omega}_2 - \overline{\Omega}_1) . \qquad (2.18a)$$

From (2.12) and (2.7), (2.18a) can be written in the form

$$\sigma = + \left[(\epsilon - \omega_0)^2 + |\Delta|^2 \right]^{1/2} . \tag{2.18b}$$

In (2.17),

$$g(\omega) = \epsilon d\omega^2 \sqrt{2\pi} / \sqrt{\omega V} , \qquad (2.19)$$

where d is the matrix element of the transition dipole moment, and we have taken

 $\gamma \approx \gamma_k$

for all modes k. It is emphasized that the integral (2.17) cannot be evaluated rigorously owing to the approximations in this model. We can, however, obtain a plausible approximate evaluation as follows in the next paragraph.

The density of modes in free space is given by

$$S(\omega) = 2\omega^2/(2\pi)^3$$
. (2.20)

If we use this as an analogy, then (2.17) can be written in the form

$$I = 2\pi\epsilon^2 d^2 V \int_0^\infty d\omega \int_{4\pi} d\Omega \frac{S(\omega)}{\omega} \times \left(\frac{1}{\omega - \omega_0 - \sigma + i\gamma} + \frac{1}{\omega - \omega_0 - \sigma - i\gamma}\right).$$
(2.21)

If we assume that all the cavity modes are well separated, then

$$VS(\omega) = s_0 \delta(\omega - \omega_c), \qquad (2.22)$$

where ω_c is the center frequency of a principal cavity mode which interacts with the atomic system and s_0 is the total number of cavity modes in the neighborhood of ω_c . Thus

$$\int_{4^{\pi}} d\Omega S(\omega) = K\delta(\omega - \omega_c), \qquad (2.23a)$$

where

 $K = s_0 / V \tag{2.23b}$

is the density of cavity modes in the neighborhood of the principal cavity mode. If (2.23) is used in (2.21), the result is

$$I = (g_0^2/N)(\omega_c - \omega_0 - \sigma)/[(\omega_c - \omega_0 - \sigma)^2 + \gamma^2],$$
(2.24)

where

 $g_{0}^{2} = [(4\pi)^{2} d^{2} K / \omega_{c}] \overline{\rho}$ (2.25)

and

$$\overline{\rho} = N/\lambda_r^3, \quad \lambda_r = 2\pi/\epsilon . \tag{2.26}$$

The "density" $\overline{\rho}$ is the number of atoms within a cubic wavelength λ_r at atomic resonance.

The expression (2.24) is sensitive to detuning of the laser frequency ω_0 from the atomic baretransition frequency ϵ [(2.18b)] as well as detuning of the cavity frequency ω_c from the laser frequency. It should be emphasized that (2.24) is derived from a simple model. For example, in the case of perfect tuning, $\omega_c = \omega_0 + \sigma$, other cavity modes should contribute, and *I* is not zero. Also, the atoms are assumed to be fixed in position. This expression, which contains both absorptive and dispersive contributions, will be discussed in later sections.

We can now define an effective atomic dipoledipole coupling \overline{g} which arises entirely from the atom-atom interaction via the radiation field:

$$\overline{g} \equiv NI . \tag{2.27}$$

If we use (2.16) and the definition (2.27) in (2.15), we obtain the effective atomic interaction Hamiltonian

$$H'_{T} = -\frac{\overline{g}}{N} \sum_{\substack{j, l=1\\ j \neq l}}^{N} C_{2l}^{\dagger} C_{1l} C_{1j}^{\dagger} C_{2j} + \left(\alpha^{*} \sum_{j=1}^{N} C_{1j}^{\dagger} C_{2j} + \text{H.c.} \right) .$$
(2.28)

Equation (2.28) is the main result of this section, and will be the working interaction Hamiltonian in the calculations which follow. It is to be noted that \overline{g} is a function of the internal field via (2.11), (2.12), (2.18b), and (2.24). This is not surprising, since, as pointed out above, it arises entirely from the atom-atom interaction via the internal radiation field.

In Sec. III the thermodynamic Green's functions and associated correlation functions pertinent to the determination of the macroscopic atomic polarization in thermodynamic equilibrium in the rotating frame are defined and the condition for bistable behavior is derived.

III. FIRST-ORDER PHASE TRANSITION AND OPTICAL BISTABILITY

For the purpose of calculating the macroscopic transverse polarization in thermodynamic equilibrium, the following set of single-particle Green's functions is defined^{24,27}:

$$G_{n,i}(t-t') = -i\,\theta(t-t')\langle [C_{n,i}(t), C_{n,i}^{\dagger}(t')] \rangle , \qquad (3.1)$$

$$F_{1,i}(t-t') = -i\,\theta(t-t') \langle [C_{2,i}(t), C_{1,i}^{\dagger}(t')] \rangle , \qquad (3.2)$$

$$F_{2,i}(t-t') = -i\,\theta(t-t')\langle [C_{1,i}(t), C_{2,i}^{\dagger}(t')] \rangle , \qquad (3.3)$$

where θ is the unit step function, i.e.,

$$\theta(\tau) = 1, \quad \tau > 0,
\theta(\tau) = 0, \quad \tau < 0,$$
(3.4)

and n in (3.1) is either 1 or 2. It is also expedient to define the time-independent correlation functions

$$\hat{F}_{1,i} = \langle C_{1,i}^{\dagger}(t) C_{2,i}(t) \rangle, \qquad (3.5)$$

$$\hat{F}_{2,i} = \langle C_{2,i}^{\dagger}(t) C_{1,i}(t) \rangle .$$
(3.6)

In expressions (3.1)-(3.6) and in what follows, $\langle \rangle$ is the thermal average. These averages are with respect to the rotating frame. Thus the density operator ρ in this representation is assumed to be an exponential of the form

$$\rho \alpha \, e^{-\beta_S H_T}, \tag{3.7}$$

where

$$\beta_s = 1/kT_s \tag{3.8}$$

and T_s is an effective temperature, commonly referred to as the "spin temperature" in the rotating frame.²⁸ Therefore, for any operator A referred to the rotating frame, its thermal average is explicitly

$$\langle A \rangle = \operatorname{tr}(A \ e^{-\beta_{s}\hat{H}} T)/Z,$$
 (3.9a)

$$Z = \operatorname{tr} e^{-\beta_{S} H_{T}}, \qquad (3.9b)$$

where

$$\hat{H}_T = H_T - \mu N \tag{3.10}$$

and μ is the chemical potential.

If the effective atomic interaction Hamiltonian (2.28), together with (2.6a), is used to calculate the Heisenberg equations of motion for the $C_{n,j}$, and if the decoupling scheme

$$\langle [C_{1,j}^{\dagger}(t)C_{2,j}(t)C_{1,i}(t), C_{1,i}^{\dagger}(t')] \rangle \approx \langle C_{1,j}^{\dagger}(t)C_{2,j}(t) \rangle \langle [C_{1,i}(t), C_{1,i}^{\dagger}(t')] \rangle$$

$$(3.11)$$

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is employed, the coupled equations of motion for the Green's functions (3.1) (n = 1), and (3.2) are

$$\left(i\frac{\partial}{\partial t} - \frac{1}{2}\Omega_1\right)G_1(t-t') = \delta(t-t') - (\overline{g}\widehat{F}_2 - \alpha^*)F_1(t-t'),$$
(3.12)

$$\left(i\frac{\partial}{\partial t}-\frac{1}{2}\Omega_2\right)F_1(t-t')=-(\bar{g}\hat{F}_1-\alpha)G_1(t-t')\,,\quad(3.13)$$

where the single-particle index has been dropped consistent with the assumption that all the atoms are identical.

The decoupling approximation (3.11) amounts to ignoring combinations of atomic operators which lead to non-energy-conserving averages, and to the assumption that operator products involving different atoms can be factorized. The latter assumption is explicitly what we mean by the "mean-field" approximation. The mean-field approximation is certainly justified in our model, which assumes that all of the atoms are contained in a volume much smaller than an atom resonance wavelength. It should be noted that our mean-field approximation is quite different in principle to that of Refs. 7 and 8, but identical to that of Refs. 10 and 19.

The coupled partial differential equations (3.12)and (3.13) form a closed set and can be easily solved algebraically by Fourier-transforming the equations. The transformed equations are

$$(\omega - \frac{1}{2}\Omega_1)G(\omega) = 1/2\pi - \Delta F(\omega), \qquad (3.14)$$

$$(\omega - \frac{1}{2}\Omega_2)F(\omega) = -\Delta^* G(\omega), \qquad (3.15)$$

where

$$\Delta \equiv \overline{g} \widetilde{F}_2 - \alpha^* \,. \tag{3.16}$$

We have now dropped the subscripts on the Green's functions F_1 and G_1 , since Eqs. (3.12) and (3.13) form a closed set. A similar set of equations is obtained for F_2 and G_2 .

The expressions (3.14) and (3.15) can be combined to give

$$\left[\left(\omega - \frac{1}{2} \Omega_1 \right) \left(\omega - \frac{1}{2} \Omega_2 \right) - |\Delta|^2 \right] F(\omega) = -\Delta^* / 2\pi.$$

(3.17)

This last equation can be written more tractably in the form

$$\left[(\omega - \Gamma_{+})(\omega - \Gamma_{-}) \right] F(\omega) = -\Delta^{*}/2\pi, \qquad (3.18)$$

where Γ_{\star} and Γ_{-} are the roots of

$$(\omega - \frac{1}{2}\Omega_1)(\omega - \frac{1}{2}\Omega_2) - |\Delta|^2 = 0.$$
 (3.19)

Explicitly,

$$\Gamma_{\pm} = \pm \frac{1}{2} \left[(\epsilon - \omega_0)^2 + |\Delta|^2 \right]^{1/2} . \tag{3.20}$$

The spectral theorem for thermodynamic Green's functions²⁷ is now used to calculate the macroscopic transverse polarization \mathcal{F} :

$$\mathfrak{F} \equiv \langle C_1^{\dagger} C_2 \rangle , \qquad (3.21a)$$

$$\mathfrak{F} = \hat{F}_1 = \hat{F}_2^* \,. \tag{3.21b}$$

We have, from the spectral relation,²⁷

$$\mathfrak{F} = i \lim_{\delta \to 0} \int_{-\infty}^{\infty} \frac{F(\omega + i\delta) - F(\omega - i\delta)}{e^{\beta_{\mathcal{S}}(\omega - \mu)} - 1} d\omega . \qquad (3.22)$$

Consistent with the assumption in the model that the wave functions of the atoms do not overlap, the quantum statistics reduce to classical statistics under the assumption that the chemical potential μ and β_s satisfy²⁹

$$e^{-\beta_s \mu} \to \infty \tag{3.23}$$

in the rotating frame. From the condition for conservation of particle number,

$$9_1 + 9_2 = 1$$
, (3.24a)

where

$$\mathbf{9}_n \equiv \langle C_n^{\dagger} C_n \rangle, \qquad (3.24b)$$

and \mathfrak{S}_n is obtained from G_n by a relation identical to (3.22), we obtain the additional condition

$$e^{\beta_{s}\mu} = \left[\left(e^{\beta_{s}\sigma/2} + e^{-\beta_{s}\sigma/2} \right) \right]^{-1}, \qquad (3.25)$$

where σ is given by (2.18b), and from (3.20)

$$\sigma = 2|\Gamma_{\pm}|. \tag{3.26}$$

It should be noted at this point that $|\Delta|$ appearing in (2.18b) is now determined by (3.16).

Thus, using (3.18) and (3.25) in (3.22), together with the condition (3.23), we can evaluate the integral in a straightforward manner, finding

$$\mathcal{F} = 2\Delta * \tanh(\frac{1}{2}\beta_s \sigma) / \sigma. \qquad (3.27)$$

It is useful to make the identification

$$E_I \equiv \overline{g} \mathfrak{F} - \alpha \tag{3.28a}$$

for the internal field, and

$$E_A \equiv -\alpha \tag{3.28b}$$

for the applied field amplitude. Then, after multiplying both sides of (3.27) by \overline{g} , we have, using (3.28),

$$E_I - E_A = 2\overline{g}E_I \tanh\frac{1}{2}\beta_s \sigma/\sigma. \qquad (3.29)$$

Equation (3.29) is the main result of this section, and may or may not lead to OB, depending upon the sign and magnitude of \overline{g} , (2.27) and (2.24), and the magnitude of the argument of the hyperbolic tangent. It is emphasized that β_s corresponds to an effective temperature which is the exact analog of a spin temperature in the rotating

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frame.²⁸ The relationship between T_s and T is discussed in Ref. 28.

It is useful to define dimensionless quantities

$$x = E_I / \gamma, \quad y = E_A / \gamma,$$
 (3.30a)

$$\delta = (\omega_c - \omega_0)/\gamma, \quad \nu = (\epsilon - \omega_0)/\gamma, \quad (3.30b)$$

$$C = g_0^2 / \gamma^2$$
, (3.30c)

where g_0 is given by (2.25). With the substitutions (3.30), (3.29) has the form

$$y = x - 2C \frac{(\delta - \sigma')x}{(\delta - \sigma')^2 + 1} \frac{\tanh \frac{1}{2}\beta_s \gamma \sigma'}{\sigma'}, \qquad (3.31)$$

where

$$\sigma' = + (\nu^2 + x^2)^{1/2} . \tag{3.32}$$

In the low-spin-temperature regime, which is implicitly assumed in the models presented in Refs. 5-10,

$$\tanh \frac{1}{2}\beta_s \gamma \sigma' \xrightarrow[T_s \to 0]{} 1,$$

and (3.31) becomes

$$y = x - \frac{2C(\delta - \sigma')x}{[(\delta - \sigma')^2 + 1]\sigma'}.$$
 (3.33)

If the cavity and laser are perfectly tuned, i.e.,

 $\delta = \nu = 0,$

then (3.33) reduces to

$$y = x + 2Cx/(1+x^2).$$
 (3.34)

This relation is identical in form with the corresponding relation obtained by BL in Ref. 7 for the relation between the input and output fields of a ring cavity loaded with a saturable absorber far from thermodynamic equilibrium. We would, in fact, expect to get reasonable agreement between the two models in the low-temperature limit.

It is well known that OB exists^{7,8} for the field y as a function of the field x in (3.34), if C > 4. For values of the system constant C for which C < 4, x is a monotonic function of y and there is no OB.

Equation (3.33) shows the effect of cavity detuning δ and laser detuning ν [Eq. (3.30b)]. This expression contains dispersive as well as absorptive contributions. Figure 1 shows the equilibrium values for the input y as a function of the cavity field x, according to (3.33) for various equal values of δ and ν and for C = 40. It is seen that absorption, i.e., $\delta = \nu = 0$ results in the maximum value for the initial turning point and largest bistable effect, whereas for larger values for δ and ν , the turning point occurs at smaller input y and the OB effect becomes smaller.



FIG. 1. Effect of cavity and laser detuning on optical bistability. Input field y vs internal field x from (3.33). Curve a: C=40, $\delta=\nu=0$; curve b: C=40, $\delta=\nu=2$; curve c: C=40, $\delta=\nu=4$.



FIG. 2. Asymmetric effect of cavity detuning on optical bistability. Input field y vs internal field x from (3.33). Curve a: C = 60, $\delta = \nu = 10$; curve b: same parameters as for curve a, except $\delta = -9$; curve c: same parameters as for curve a, except $\delta = 9$; curve d: same parameters as for curve a, except $\delta = 8$.

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Figure 2 shows the asymmetric behavior of cavity detuning δ for a fixed laser detuning ν from the atomic frequency, according to (3.33). For the four curves in this figure C = 60 and $\nu = 10$. The curves labeled c and b have the same values for the parameters, except that $\delta = 9$ for c and -9 for b. Curve a corresponds to the maximum turning point and largest OB, and has $\delta = \nu$. As δ decreases in value and remains positive, the input value of the first turning point decreases as well as the OB effect. Curve c represents approximately the condition for infinite differential gain (threshold) whereas for smaller values of δ (curve d), there is no OB. The asymmetric behavior with cavity detuning for fixed laser off tuning from the atomic transition depicted in Fig. 2 appears to be consistent with the experimental results of Gibbs, McCall, and Venkatesan.⁵

The decrease in the value of the input y for the first turning point with off tuning, shown in Figs. 1 and 2, is not in agreement with the results of the absorption-dispersion model of BL¹⁸ and Hassan et al.¹⁹ The opposite effect occurs in the results of their models. We do not at present have an explanation for this discrepancy. Our model, owing to the physical assumptions made. is, however, physically different than that of Refs. 18 and 19. In the case of the latter, the atomic system is treated as always far from thermodynamic equilibrium, whereas in our model, the system is characterized by a spin temperature and is well below laser threshold. Where dispersive effects are dominant, it may be better to represent the system as in a state near thermodynamic equilibrium rather than far removed from equilibrium.

The expression (3.31) describes the conditions for OB and has the form of a first-order phase transition. The phase transition corresponds to spontaneous ordering of the atomic dipoles, coupled via the light field, to produce a macroscopic dipole moment. This stems from the particular form for the effective atomic interaction (2.28), (2.27), and (2.24). In Sec. IV we shall discuss the connection between the first-order phase transition described by (3.31) and the second-order or "super-radiant" phase transition in the limit of zero applied field.

IV. PHASE TRANSITIONS IN THE ABSENCE OF APPLIED FIELD

This section will be used to discuss the secondorder "super-radiant" phase transition¹²⁻¹⁶ in the zero-applied-field limit of (3.31).

Up to this point, the thermodynamics which we employed have been relative to the coordinate system rotating at the applied-field carrier frequency ω_0 . Here, we wish to examine the properties of (3.31) in the limit

$$\omega_0 \neq 0, \qquad (4.1)$$

$$y=0. (4.2)$$

For these conditions, (3.8) becomes

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$$T_s \to T = (k\beta)^{-1}, \qquad (4.3)$$

where T is the temperature of the surroundings with which our system is in temperature equilibrium under new conditions (4.1) and (4.2).

For these circumstances, (3.31) can be written in the form

$$\Delta^* \left(1 - \frac{2g_0^2}{\epsilon} \frac{(\omega_c - \epsilon \eta)}{(\omega_c - \epsilon \eta)^2 + \gamma^2} \frac{\tanh \frac{1}{2}\beta\epsilon\eta}{\eta} \right) = 0. \quad (4.4)$$

Here,

$$\eta = + (1 + |\Delta|^2 / \epsilon^2)^{1/2} , \qquad (4.5)$$

and the order parameter Δ (macroscopic transverse polarization) is given by (3.16) with α^* set equal to zero. Expression (4.4) is satisfied for all values of the temperature *T* if either

$$\Delta^* = 0 \quad (disordered phase) \tag{4.6}$$

 \mathbf{or}

$$\frac{\tanh \frac{1}{2}\beta\epsilon\eta}{\eta} = \frac{\epsilon}{2g_0^2} \frac{(\omega_c - \epsilon\eta)^2 + \gamma^2}{\omega_c - \epsilon\eta} \quad \text{(ordered phase).}$$
(4.7)

If the last equation can be satisfied in the limit $\Delta \rightarrow 0$, then, in this limit, it defines a critical temperature T_c below which $\Delta \neq 0$ (ordered phase) and above which $\Delta = 0$ (disordered phase).

In the limit $\Delta \rightarrow 0$, i.e., $\eta \rightarrow 1$, (4.7) becomes

$$\tanh \frac{1}{2}\beta_c \,\epsilon = \frac{\epsilon}{2g_0^2} \frac{(\omega_c - \epsilon)^2 + \gamma^2}{\omega_c - \epsilon} \,. \tag{4.8}$$

Immediately, it is seen that this expression differs markedly from the conditions for the existence of the super-radiant phase transition discussed in earlier works.¹²⁻¹⁷ In particular, (4.8) depends upon the cavity detuning $\omega_c - \epsilon$, which does not appear in the earlier works. This discrepancy stems explicitly from the particular form for the effective coupling \overline{g} , (2.27) and (2.24), which arises because of the specific conditions imposed in this model. In the present case, \overline{g} is specialized according to specific characteristics of the cavity, i.e., ω_c and γ , and the form which it takes under these cavity conditions, arises because of the integration over all modes [(2.17)].

In summary, the condition that (4.8) be satisfied, and a critical temperature defined, is that

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$$(\omega_c - \epsilon)^2 + \gamma^2 \le (2g_0^2/\epsilon)(\omega_c - \epsilon).$$
(4.9)

If $(\omega_c - \epsilon) \neq 0$, (4.8) determines a finite critical temperature T_c , and (4.7) determines values of the order parameter η for temperatures $T < T_c$. If $y \neq 0$ in (3.31), there is no second-order phase transition.

Our work has brought OB of Eq. (3.31) and the super-radiant phase transition Eq. (4.4) to the same footing. Equation (4.9) can be written as

$$\frac{\omega_{c}-\epsilon}{\left[(\omega_{c}-\epsilon)^{2}+\gamma^{2}\right]^{1/2}}\frac{C}{\epsilon/\gamma}\frac{1}{(\omega_{c}-\epsilon)/\gamma} \ge 1.$$
(4.10)

If the observation of OB suggests the order of magnitude of C and $(\omega_c - \epsilon)/\gamma$, i.e., $C \ge 1$ and $(\omega_c - \epsilon)/\gamma \approx 1$, then (4.10) shows that ϵ/γ must be of the same order as $(\omega_c - \epsilon)/\gamma$ for observation of the second-order phase transition. This is a condition not satisfied experimentally. Thus OB and the lack of experimental evidence of the super-radiant phase transition are consistent with the results of our model.

V. CONCLUSIONS

The results of this work contain, for the first time, all of the qualitative features of optical bistability⁵ from a purely thermodynamic point of view [Eq. (3.31) and Figs. 1 and 2]. Equation (3.31) provides a description of OB as a firstorder phase transition in the light-matter interaction (2.2b), where it was shown that the ordering process stems explicitly from dipole-dipole pair correlation (2.28) via the mutual internal radiation field. The form for the dipole-dipole interaction (2.28) is identical to the retarded dipole-dipole interaction that gives rise to cooperative radiation reaction and frequency shifts in superfluorescence,^{25,26} and is the same effective interaction responsible for the existence of the "super-radiant" phase transition in thermodynamic equilibrium.^{12-16,24} The interaction is also identical in form to the interaction coupling Cooper pairs in the Bardeen-Cooper-Schrieffer theory of ${\tt superconductivity.}^{22-24}$

Our results not only give the conditions for OB in terms of characteristic material and cavity parameters, but also incorporate the effects of dispersion as well as absorption [(3.31)]. The effects of cavity and laser detuning arise explicitly from the effective coupling (2.27) and (2.24) in the atom-field interaction. These results, (3.31), qualitatively predict the observed⁵ asymmetry in OB with respect to cavity detuning which arises because of dispersive contributions (Fig. 2). Our results agree with the results of other models for OB^{7-10,17} in the low-temperature, absorption limit. The results we derived differ from the results of other models in the dispersive contributions^{18,19} and also from the fact that our results are characterized by the exact analog (3.31) of a "spin" temperature.²⁸

In addition to the establishment of OB as a first-order phase transition [(3.31)] in the presence of an external field, we have shown the relationship, using the same model, to the super-radiant second-order phase transition in the absence of the applied field [(4.7) and (4.8)]. The condition for the super-radiant phase transition is characterized by a critical temperature (4.8) and an order parameter (4.5), which corresponds to the existence of a nonzero macroscopic transverse polarization in the material for $\eta > 1$. The existence of the second-order phase transition [(4.8)] and the behavior of the order parameter at temperatures near the critical temperature [(4.7)] is governed not only by the material parameters, but also by the cavity frequency ω_c , and mirror reflectivity contained in the photon escape rate γ [(4.9)]. As discussed in Sec. IV, introduction of an external laser field removes the conditions [(4.7) and (4.8)] for a second-order phase transition and replaces them with a condition for a first-order OB phase transition, (3.31).

The free energy as a function of temperature and order parameter for zero and nonzero values of the applied field and its behavior in the neighborhood of instabilities, together with the photon statistics, will be dealt with in another publication.³⁰ The photon statistics in our model in the presence of an applied field are expected to be quite different from those of earlier models.7-10 In regard to the latter, the "upper" state of bistability is the chaotic state, i.e., the system is near laser threshold, and therefore large quantum fluctuations and resonance-fluorescence-like line shapes are expected.⁸ Our model assumes that the system is always well below laser threshold and predicts a more ordered state for the "upper" bistability state than for the lower. i.e., bistability behavior is characterized by the onset of large macroscopic polarization. The photon statistics, line shapes and behavior in the critical region are expected to be markedly different.

Finally, it should be noted that the first-order phase transition in the presence of an external field discussed in Sec. III, has been studied experimentally (OB).⁵ No such studies have been reported in connection with the phase transition in the absence of an external field discussed in Sec. IV. This fact seems to be consistent within the framework of our model as discussed at the end of the last section. The authors wish to acknowledge useful discussions with Dr. J. G. Castle and to express

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