

Equilibrium Statistical Mechanics of Matter Interacting with the Quantized Radiation Field

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(Received 7 June 1973)

The thermodynamic properties of several systems of multilevel atoms interacting with a quantized radiation field are investigated. We allow a quantum-mechanical treatment of the translational degrees of freedom and do not require the rotating-wave approximation. In the finite-photon-mode case one can calculate the free energy per atom in the thermodynamic limit exactly and rigorously. In the infinite-mode case we only get upper and lower bounds, but these are sufficient to give conditions for thermodynamic stability and instability. The kind of phase transition previously found by us for the one-mode Dicke model with the rotating-wave approximation persists in the general multimode case.

I. INTRODUCTION

In a previous paper¹ we elucidated the thermodynamic properties of the Dicke maser model with one photon mode coupled to N two-level atoms in the rotating-wave approximation. We calculated the free energy per atom and the thermal expectation values of time-dependent intensive and fluctuation observables exactly in the limit $N \rightarrow \infty$. Our analysis relied heavily on the various conservation laws inherent in the model: the total "number" operator and the total atomic "spin." In the relevant part of the spectrum we effectively diagonalized the Hamiltonian in such a way that the error became negligible as $N \rightarrow \infty$. While such a procedure is necessary if one wants to study fluctuation observables, it is intuitively clear that a simpler mean field method must exist if one is content to analyze only the intensive observables. In fact our results showed that these quantities behave classically; that is to say the lack of commutativity becomes unimportant as $N \rightarrow \infty$. It is also clear that these qualitative features should continue to hold even without the above mentioned conservation laws, i.e., it should be possible to handle a much wider class of models. In this paper we show how to achieve that goal in a simple way by using coherent states and the results of a recent paper.² The phase transition found in Ref. 1 persists in the general multimode case.

Wang and Hioe³ and Hioe⁴ demonstrated that the Glauber coherent states of the photon field constitute a natural basis for this problem. Unfortunately, they were not able to show that their approximate evaluation of $N^{-1} \ln \text{Tr} e^{-\beta H}$ is exact when $N \rightarrow \infty$. Nevertheless, it is a fact that their

answer agreed precisely with our previous rigorous result¹ for the Dicke model, and we shall prove here that their results for the other models they treat are also correct. In addition, we also calculate exactly the expectation values of intensive observables for those models. Ginibre⁵ has previously made use of coherent states to obtain exact results for the many-boson problem. Our methods can be easily generalized for atoms of more than two levels.

In Sec. III we make an attempt to understand the thermodynamics of the infinite-mode case which, to the best of our knowledge, was never discussed before. We cannot solve that problem exactly, but we can derive upper and lower bounds to the partition function which prove that such a system is thermodynamically stable if and only if the atoms have very repulsive cores.

In Sec. IV we return to the finite-mode Hamiltonian, but allow translational degrees of freedom (either quantum or classical) for the atoms. It is shown there that use of the Glauber coherent photon states permits an exact (as $N \rightarrow \infty$) reduction of the problem to a conventional many body problem. This, in turn, can be solved in closed form in several interesting cases.

We do not confine our attention only to the Glauber coherent photon states, but also show that the Bloch atomic coherent states⁶ for the atoms may be used to advantage. The former make the radiation field classical, while the latter make the atomic variables classical. It is the Bloch states that are used in our discussion of the infinite-mode case.

In the Appendix we resolve some technical problems related to the unbounded nature of the photon operators.

II. FINITE-MODE MODELS

The Hamiltonians we shall consider are those of the form

$$H = \sum_{m=1}^M \nu_m a_m^* a_m + \epsilon S^z + N^{-1/2} \sum_{m=1}^M \sum_{n=1}^N [a_m (\lambda_{mn} S_n^+ + \mu_{mn} S_n^-) + \text{H.c.}], \quad (2.1)$$

where a_1^*, \dots, a_M^* are boson creation and annihilation operators for the photon modes having energies $\nu_m > 0$, $\tilde{S}_1, \dots, \tilde{S}_N$ are spin- $\frac{1}{2}$ operators describing N two-level atoms having energy spacing $\epsilon > 0$ ($S_n^+ = S_n^x + iS_n^y$ and $S_n^- = S_n^x - iS_n^y$, $i = x, y$ or z). The λ_{mn} (respectively, μ_{mn}) are coupling constants for the rotating (respectively, counter-rotating) wave terms. The factor $N^{-1/2}$ in (2.1) really comes from a factor $V^{-1/2}$, where V is the volume of the cavity containing the atoms. However, as we are interested in N/V fixed, the distinction merely entails a coupling constant renormalization. The interaction term in (2.1) is linear in the a^* operators, but the method we are about to describe would work equally well if one included quadratic terms (i.e., if one goes beyond the dipolar approximation).

A. Atomic Coherent State Representation

We turn now to the atomic or Bloch angular momentum coherent states⁶ used in Ref. 2. With \mathcal{F} being the Fock space for all the photon modes and \mathcal{K} being the Hilbert space for the N atoms (spins) we define

$$Z = \text{Tr} e^{-\beta H} \quad (2.2)$$

where the trace is over $\mathcal{F} \otimes \mathcal{K}$. The bounds obtained in Ref. 2, and justified in the Appendix for boson operators, are

$$\tilde{Z}(\tfrac{1}{2}) \leq Z \leq \tilde{Z}(\tfrac{3}{2}), \quad (2.3)$$

where

$$\tilde{Z}(J) = 2^N \text{Tr}_{\mathcal{F}} (4\pi)^{-N} \int d\Omega^N e^{-\beta \tilde{H}(J, \Omega^N)} \quad (2.4)$$

and where

$$\Omega^N = (\tilde{\Omega}_1, \dots, \tilde{\Omega}_N),$$

$$\tilde{\Omega}_n = (\sin \theta_n \cos \varphi_n, \sin \theta_n \sin \varphi_n, \cos \theta_n). \quad (2.5)$$

$\tilde{H}(J, \Omega^N)$ is defined from (2.1) by replacing each \tilde{S}_n by $J\tilde{\Omega}_n$. The bounds in (2.3) for the free energy per atom do not agree as $N \rightarrow \infty$, but they are useful in proving the stability of the infinite photon mode free energy—a subject to which we shall return in Sec. III.

Further progress can be made with the Bloch-state bounds, however, if we make an assumption about H , namely that the λ_{mn} and the μ_{mn} are independent of n . This is true in the original Dicke model where one neglects the spatial variation of the radiation field. (Alternatively, the method will work if the atoms belong to a finite number of groups in each of which the coupling constants are the same.) In this case we can use the fact that the total “spin,” J , is a constant of the motion, so that

$$Z = \sum_{J=0}^{N/2} Y(N, J) Z(J), \quad (2.6)$$

$$Z(J) = \text{Tr} e^{-\beta H(J)}, \quad (2.7)$$

where $H(J)$ is the Hamiltonian for a spin, \tilde{S} , of magnitude J ,

$$H(J) = \sum_{m=1}^M \nu_m a_m^* a_m + \epsilon S^z + N^{-1/2} \sum_{m=1}^M [a_m (\lambda_m S^+ + \mu_m S^-) + \text{H.c.}] \quad (2.8)$$

and

$$Y(N, J) = N! (2J+1)! [(J+1 + \tfrac{1}{2}N)! (\tfrac{1}{2}N - J)!]^{-1} \quad (2.9)$$

is the number of ways to construct an angular momentum J from N spin- $\frac{1}{2}$ particles.

Using Ref. 2, one has the bounds

$$\tilde{Z}(J) \leq (2J+1)^{-1} Z(J) \leq \tilde{Z}(J+1), \quad (2.10)$$

with

$$\tilde{Z}(J) = (4\pi)^{-1} \text{Tr}_{\mathcal{F}} \int d\Omega e^{-\beta \tilde{H}(J, \Omega)} \quad (2.11)$$

and $\tilde{H}(J, \Omega)$ is (2.8) with S replaced by $J\tilde{\Omega}$. Since \tilde{H} is quadratic in the a_m^* , it is easy to evaluate $\text{Tr}_{\mathcal{F}}$ in (2.11):

$$\tilde{Z}(J) = (4\pi)^{-1} \prod_{m=1}^M (1 - e^{-\beta \nu_m})^{-1} \int_0^\pi \sin \theta d\theta \int_0^{2\pi} d\varphi \exp \left(-\beta \epsilon J \cos \theta + \beta N^{-1/2} J^2 \sin^2 \theta \sum_{m=1}^M |\lambda_m e^{i\varphi} + \mu_m e^{-i\varphi}|^2 \nu_m^{-1} \right). \quad (2.12)$$

Since $J^2 \leq \frac{1}{4}N^2$, we see that

$$\ln \tilde{Z}(J+1) - \ln \tilde{Z}(J) \leq \beta \epsilon + \beta [(N+1)/N] \sum_{m=1}^M (|\lambda_m| + |\mu_m|)^2 \nu_m^{-1}, \quad (2.13)$$

which means that (2.12) gives the free energy per atom exactly to order M/N . It is also clear that for large N and fixed M , (2.12) and (2.6) can be evaluated by steepest descent, i.e., simply by maximizing with respect to θ , φ , and J . If we first maximize with respect to φ we see that *the free energy is the same as that for one mode in the rotating-wave approximation*, but with an effective coupling constant λ given by

$$\lambda^2 = \max_{\varphi} \sum_{m=1}^M |\lambda_m e^{i\varphi} + \mu_m e^{-i\varphi}|^2 \nu_m^{-1}. \quad (2.14)$$

Subsequent maximization with respect to θ and J yields the free energy found in Ref. 1. Such a result agrees with that found in Ref. 3.

Expectation values of intensive observables can be found by the foregoing procedure, but some technicalities are required. These will be discussed in the Appendix. The result is as expected: The expectation value of any polynomial in the intensive observables (defined in the Appendix) is equal to that polynomial evaluated at the maximal steepest descent points referred to above, and then averaged over the variety of maximal steepest descent points if there is more than one point.

Before turning to the photon coherent states, which we shall describe next, we remark that when there is no spatial dependence (λ_{mn} and μ_{mn} independent of n) the Bloch picture (2.12) is the more convenient to use. The main conclusion, (2.14), that even without the rotating-wave approximation there is effectively only one coupling constant, is more tedious to derive using the photon coherent states (cf. Refs. 3 and 4).

B. Photon Coherent-State Representation

For a given photon mode, $a^\#$, a Glauber coherent state is

$$|\alpha\rangle = \exp[-\frac{1}{2}|\alpha|^2 + \alpha a^\#] |0\rangle, \quad (2.15)$$

where α is any complex number and $|0\rangle$ is the photon vacuum. The following properties are required and are justified in the Appendix:

$$\langle\alpha|\beta\rangle = \exp[-\frac{1}{2}|\alpha|^2 - \frac{1}{2}|\beta|^2 + \alpha^*\beta], \quad (2.16)$$

$$\text{Tr}|\alpha\rangle\langle\beta| = \langle\beta|\alpha\rangle, \quad (2.17)$$

$$\text{Tr}A = \pi^{-1} \int d\alpha \langle\alpha|A|\alpha\rangle, \quad (2.18)$$

where A is any trace-class operator and $\int d\alpha = \int_{-\infty}^{\infty} d(\text{Re}\alpha) d(\text{Im}\alpha)$,

$$\langle\alpha|a|\alpha\rangle = \alpha, \quad (2.19)$$

$$\langle\alpha|a^*a|\alpha\rangle = |\alpha|^2, \quad (2.20)$$

$$I = \pi^{-1} \int d\alpha |\alpha\rangle\langle\alpha|, \quad (2.21)$$

$$a = \pi^{-1} \int d\alpha \alpha |\alpha\rangle\langle\alpha|, \quad (2.22)$$

$$a^*a = \pi^{-1} \int d\alpha (|\alpha|^2 - 1) |\alpha\rangle\langle\alpha|. \quad (2.23)$$

The same methods as in Ref. 2, but using the Glauber states instead of the Bloch states, yield upper and lower bounds for the partition function (again, see the Appendix):

$$\bar{Z} \leq Z \leq \exp\left(\beta \sum_{m=1}^M \nu_m\right) \bar{Z}, \quad (2.24)$$

where

$$\bar{Z} = \pi^{-M} \text{Tr}_{\mathcal{H}} \int d\alpha^M e^{-\beta \bar{H}(\alpha^M)}, \quad (2.25)$$

and

$$\begin{aligned} \alpha^M &= (\alpha_1, \dots, \alpha_M), \\ \bar{H}(\alpha^M) &= \sum_{m=1}^M \nu_m |\alpha_m|^2 + \epsilon S^z + N^{-1/2} \\ &\times \sum_{m=1}^M \sum_{n=1}^N [\alpha_m (\lambda_{mn} S_n^+ + \mu_{mn} S_n^-) + \text{H.c.}]. \end{aligned} \quad (2.26)$$

The trace on \mathcal{H} in (2.25) is now easy to compute:

$$\begin{aligned} \bar{Z} &= \pi^{-M} \int d\alpha^M \exp\left(-\beta \sum_{m=1}^M \nu_m |\alpha_m|^2\right) 2^N \prod_{n=1}^N \cosh \\ &\times \left[\frac{1}{2} \beta \left(\epsilon^2 + 4N^{-1} \left| \sum_{m=1}^M (\alpha_m \lambda_{mn} + \alpha_m^* \mu_{mn}^*) \right|^2 \right)^{1/2} \right]. \end{aligned} \quad (2.27)$$

This \bar{Z} is precisely the approximate partition function derived by Wang and Hioe.^{3,4} We note from (2.24) that, as long as the number of photon modes is finite, $-\beta N^{-1} \ln \bar{Z}$ is the free energy per atom to order MN^{-1} . Another remark is that although we assumed each atom to have only two levels, the atomic trace in (2.26) can just as well be evaluated for multilevel atoms, and the interaction does not have to be linear in the atomic S^\pm operators.

III. THERMODYNAMIC STABILITY OF THE INFINITE-MODE SYSTEM

In this section we return to the Hamiltonian H of (2.1) and shall show that under suitable conditions it is stable when the number of photon modes is infinite. This means that there exists a constant A such that $Z = \text{Tr} e^{-\beta H} < e^{NA}$ for all N . The proof of the existence of the thermodynamic limit, $\lim_{N \rightarrow \infty} N^{-1} \ln Z$, is a more complicated question with which we shall not deal. The phrase infinite number of modes means that in any frequency interval (ν_1, ν_2) the number of modes with $\nu_1 < \nu < \nu_2$

is proportional to N , the size of the cavity. The problem we are considering here is similar to the corresponding problem for electron-phonon interactions dealt with by Gallavotti, Ginibre, and Velo.⁷

We use the bounds (2.3) and the definition (2.4). The trace over the photon modes \mathcal{F} can easily be done exactly and one finds, dropping irrelevant factors of the form $e^{\text{const } N}$,

$$Z(J) \sim R_N \int d\Omega^N \exp \left(\beta J^2 \sum_{n=1}^N \sum_{j=1}^N v(\vec{\Omega}_n, \vec{\Omega}_j) - \beta \epsilon J \sum_{n=1}^N \cos \theta_n \right), \quad (3.1)$$

where

$$v(\vec{\Omega}_n, \vec{\Omega}_j) = N^{-1} \sum_m \nu_m^{-1} \text{Re}(\lambda_{mn} S_n^+ + \mu_{mn} S_n^-) \times (\lambda_{mj}^* S_j^- + \mu_{mj}^* S_j^+), \quad (3.2)$$

$$S_n^\pm = \sin \theta_n e^{\pm i \varphi_n}, \quad (3.3)$$

and R_N is the ideal photon contribution to Z

$$R_N = \prod_m (1 - e^{-\beta \nu_m})^{-1}. \quad (3.4)$$

The latter is independent of the interaction and may be assumed to have a nice thermodynamic limit in a physically sensible model, e.g.,

$$\nu_m \rightarrow \nu(\vec{k}) = |\vec{k}|, \quad (3.5)$$

$\vec{k} = 2\pi N^{-1}(n_x, n_y, n_z)$, $n_i \in \mathbb{Z}$, $\vec{k} \neq 0$. We could allow several photon modes for each \vec{k} ; such a generalization does not affect the conclusions below.

If (3.2) be inserted into (3.1), one has (dropping R_N) precisely the partition function of N classical spins of magnitude J interacting with each other via a quadratic interaction and with an external magnetic field ϵ in the z direction. This is a well understood problem. If one allows all the particles to be at the same location, i.e., λ_{mn} and μ_{mn} independent of n , then $v(\vec{\Omega}_n, \vec{\Omega}_j)$ is positive when $\vec{\Omega}_n \approx \vec{\Omega}_j$, and no matter what one assumes about the various parameters, the lower bound $\tilde{Z}(\frac{1}{2})$ will behave like e^{BN^2} with $B > 0$. Thus, *no stability is possible in this case* and, as we shall show later, the introduction of the quantum mechanical uncertainty principle does not save the situation.

To describe (3.2) more concretely, we use (3.5) *et seq.*; we associate a position \vec{x}_n with each atomic index n , and suppose that

$$\lambda_{mn} \rightarrow \lambda(\vec{k}) e^{i \vec{k} \cdot \vec{x}_n}, \quad (3.6)$$

$$\mu_{mn} \rightarrow \mu(\vec{k}) e^{i \vec{k} \cdot \vec{x}_n}.$$

Then

$$v(\vec{\Omega}_n, \vec{\Omega}_j) = \sum_{i=1}^4 g^i(\vec{\Omega}_n, \vec{\Omega}_j) h^i(\vec{x}_n - \vec{x}_j), \quad (3.7)$$

where

$$h^1(\vec{x}) = N^{-1} \sum_{\vec{k}} \nu(\vec{k})^{-1} [|\lambda(\vec{k})|^2 + |\mu(\vec{k})|^2] \cos(\vec{k} \cdot \vec{x}), \quad (3.8)$$

$$g^1(\vec{\Omega}_n, \vec{\Omega}_j) = \sin \theta_n \sin \theta_j e^{i(\varphi_n - \varphi_j)}, \quad (3.9)$$

and the other pairs g^i, h^i , $i = 2, 3, 4$ are defined similarly.

Let us investigate how the generic $h(\vec{x})$ decreases as $|\vec{x}| \rightarrow \infty$. In a real system, $\lambda(\vec{k})$ and $\mu(\vec{k})$ are the Fourier transforms of functions with exponential decrease at infinity, i.e.,

$$\lambda(\vec{k}) = \int d^3x \psi_u^*(\vec{x}) \vec{e}(\vec{k}) \cdot \vec{p} \psi_l(\vec{x}) e^{i \vec{k} \cdot \vec{x}}, \quad (3.10)$$

where u and l refer to the upper and lower atomic states, \vec{e} is a polarization vector and \vec{p} is the momentum operator. Since $\lambda(\vec{0}) \neq 0$, there is an infrared problem connected with the potential (3.8). On physical grounds we do not think this difficulty is intractable but, as a somewhat painful analysis is required, we shall not try to solve this problem here. For one thing, one should certainly include the term \vec{A}^2 in the Hamiltonian, where \vec{A} is the vector potential. This introduces a positive quadratic form in the $a^\#$ operators which can be handled by our methods, just as the photon energy, $\sum \nu_m a_m^* a_m$, was. It will certainly mitigate the k^{-1} divergence in (3.8).

From now on we shall assume $\lambda(\vec{k})$ has a zero of sufficiently high order at $\vec{k} = 0$. Then

$$h^i(\vec{x}) = \sum_{n \in \mathbb{Z}^3} \tilde{h}^i(\vec{x} + nL), \quad (3.11)$$

where L is the length of the cubic cavity and $\tilde{h}(\vec{x})$ is a smooth, real function which decreases at infinity as $|\vec{x}|^{-4}$, for example. For such a potential, it is well known⁸ that the total N body potential appearing in (3.1) is stable provided the particles are not too close together, e.g., $|\vec{x}_i - \vec{x}_j| > a > 0$ for all i, j .

To generalize the above model, (2.1), we can permit the atomic coordinates $\{\vec{x}_n\}$ to be variable and replace H by $H + U(\vec{x}_1, \dots, \vec{x}_N)$, where U is some ordinary interatomic potential, e.g., a sum of pair potentials. Now the partition function involves a configurational integral on $(\mathbb{R}^3)^N$, i.e.

$$Z \sim (N!)^{-1} \int_{\mathbb{R}^N} d^3N x Z(x) e^{-\beta U(x)}, \quad (3.12)$$

where $Z(x)$ is the partition function for each fixed set of atomic positions as defined in (2.2). The bounds (2.3) still apply and we can draw the following general conclusion: If we ignore the infrared problem as above, then the total Ham-

iltonian is stable if U is sufficiently repulsive whenever two atoms get close together and if U decays sufficiently fast (i.e., faster than $|x|^{-3-\epsilon}$ for $\epsilon > 0$) at large separations. Conversely, the Hamiltonian is unstable if U does not have strongly repulsive cores.

The next level of complication is to treat the atomic coordinates quantum-mechanically, i.e., $H \rightarrow H + U + T$, where T is the N -particle kinetic energy operator. Using Bloch states we can still derive the bounds (2.3), but now

$$\tilde{Z}(J) = 2^N \text{Tr}_{\mathcal{F}} \text{Tr}_{\mathcal{G}} \int d\Omega^N \exp[-\beta \tilde{H}(J, \Omega^N) + U(x) + T], \quad (3.13)$$

where \mathcal{G} is the Hilbert space $L^2(V^N)$ and \tilde{H} is as in (2.4). Because the terms linear in $a^\#$ involve the x_n 's, it is no longer possible to do the trace over \mathcal{F} in a closed form. To circumvent this difficulty, we derive upper and lower bounds to (3.13).

Lower bound. We use the variational principle $\text{Tre}^A > e^{(\psi, A\psi)}$ for any normalized ψ . With Ω^N fixed we choose $\psi \in \mathcal{F} \otimes \mathcal{G}$ to be $\psi = \varphi \otimes \rho$. Let ρ be of the form $\rho(\tilde{x}_1, \dots, \tilde{x}_N) = \prod_{i=1}^N \rho_i(\tilde{x}_i)$ where the ρ_i are smooth functions with supports in V which are pair-wise disjoint. Since the supports are disjoint we can imagine the atoms to be either fermions or bosons without affecting any expectation values. With ρ given, we can calculate $(\psi, T\psi)$ and $(\psi, V\psi)$. Recalling that λ_{mn} depends upon \tilde{x}_n , we can calculate $\langle \lambda_{mn} \rangle = (\psi, \lambda_{mn} \psi)$, and similarly $\langle \mu_{mn} \rangle$. Now we choose $\varphi \in \mathcal{F}$ to be the ground-state eigenvector of

$$\sum \nu_m a_m^* a_m + N^{-1/2} \sum \sum \{a_m \langle \lambda_{mn} \rangle S_n^+ + \langle \mu_{mn} \rangle S_n^- + \text{H.c.}\} \quad (3.14)$$

with energy

$$-N^{-1} \sum_m \nu_m^{-1} \left| \sum_n \langle \lambda_{mn} \rangle S_n^+ + \langle \mu_{mn} \rangle S_n^- \right|^2. \quad (3.15)$$

The point of this lower bound calculation is to show that the quantum uncertainty principle does not change the previous conclusion based on classical mechanics, namely that a repulsive core is still needed in U . Without it we still have that $Z > e^{CN^2}$ for large N with $C > 0$; this can be accomplished by letting all ρ_i have support in some fixed box of size l , where l is such that the ground-state energy of (3.14) is still $O(N^2)$ and negative. Then, $(\psi, T\psi) = O(N^{5/3} l^{-2})$, and hence the ground-state energy of $H + T$ is still $O(N^2)$ and negative.

Upper bound. We can write the Hamiltonian $H + T + U$ as $H_1 + H_2$, where $H_1 = T + \frac{1}{2} \sum \nu_m a_m^* a_m$ and

$H_2 = H' + U$, and where H' is as in (2.1), except that it has a factor $\frac{1}{2}$ multiplying the photon-energy term. The kinetic energy does not appear in H_2 , and if U has the aforementioned repulsive core plus rapid fall-off properties then the factor $\frac{1}{2}$ in H' does not affect the previous conclusion: as an operator on $\mathcal{F} \otimes \mathcal{G}$, H_2 is bounded below by $-BN$ for some constant B . Thus

$$Z < e^{\beta BN} \text{Tr}_{\mathcal{F}} \text{Tr}_{\mathcal{G}} e^{-\beta H_1} < e^{AN}$$

for some constant A .

In summary, a proper quantum statistical treatment of the atomic center of mass motion does not affect the general conclusion above.

IV. FINITE-MODE CASE WITH TRANSLATIONAL DEGREES OF FREEDOM

In Sec. III we were obliged to use the Bloch atomic state picture for a simple, but fundamental reason: If one uses the Glauber state picture, one sees from (2.20) and (2.23) that the difference between the upper and lower bounds to the free energy (2.24) is precisely $\sum_{m=1}^M \nu_m$, and if the number of modes is infinite this sum generally diverges. On the other hand, if M is finite one can always use the Glauber state picture, and the difference of the bounds is $O(N^{-1})$ in the free energy per atom. With this in mind, we shall briefly consider the class of Hamiltonians mentioned at the end of Sec. III, but with M finite, i.e.,

$$K = H + U + T, \quad (4.1)$$

where K is the total Hamiltonian in the Hilbert space $\mathcal{K} = \mathcal{F} \otimes \mathcal{G} \otimes \mathcal{H}$, H is given by (2.1), U is an ordinary interatomic potential and T is the N -atom kinetic energy.

Our bounds lead to the statement that to $O(1)$ in the total free energy

$$Z \equiv \text{Tr}_{\mathcal{K}} e^{-\beta K}, \quad (4.2)$$

$$Z = \pi^{-M} \int d\alpha^M Z(\alpha), \quad (4.3)$$

and

$$Z(\alpha) = \text{Tr}_{\mathcal{F} \otimes \mathcal{G}} e^{-\beta K(\alpha)}, \quad (4.4)$$

where $K(\alpha)$ is K with a_m and a_m^* replaced by the c numbers α_m and α_m^* . By changing variables $\alpha_m \rightarrow \gamma_m N^{1/2}$, it is clear that one can use steepest descent (in the limit $N \rightarrow \infty$) to evaluate the integral (4.3). With $\gamma = (\gamma_1, \dots, \gamma_M)$, we define

$$g(\gamma) = \lim_{N \rightarrow \infty} N^{-1} \ln Z(N^{1/2} \gamma). \quad (4.5)$$

Then,

$$\lim_{N \rightarrow \infty} N^{-1} \ln Z = \max_{\gamma} g(\gamma), \quad (4.6)$$

provided the set on which the maximum occurs is of dimension less than $2M$. The existence of the limit $g(\gamma)$ can be shown by customary methods⁸ when U is reasonable. It is generally impossible to calculate $g(\gamma)$ exactly, but the point is that the problem has been reduced to a conventional problem of point particles with spin interacting via a spin independent potential U and whose spins are coupled to a static external field which may or may not have a spatial variation. If U is a sum of single particle potentials, $g(\gamma)$ can be evaluated in closed form and there will clearly be a phase transition for sufficiently large coupling constants λ_{mn}, μ_{mn} from $\gamma = 0$ to $\gamma \neq 0$ as β is varied, just as there is in the models considered in Sec. II. An example of this in which the translational degrees of freedom are treated classically is given in Ref. 4. If U is more complicated, this kind of phase transition in γ probably still persists.

V. CONCLUSION

In this paper we have attempted to show the usefulness of coherent states (both Glauber states for the photon field and Bloch atomic states for the atomic degrees of freedom) to calculate rigorously the free energy per atom for various models of the interaction of matter and radiation. For the finite mode case (Sec. II) the problem can be reduced to the calculation of a classical type integral which can be handled by steepest descent methods. This proves the results of Hioe and Wang, and the Glauber state picture shows that, in effect, each atom interacts with a classical radiation field whose value is determined self-consistently. Therefore, one sees that as long as the number of photon modes is held fixed and as long as the interactions of the photons and atoms has a smooth spatial dependence, the thermodynamic properties of the model are very insensitive to the details of the Hamiltonian. In particular they are insensitive to whether or not the rotating-wave approximation is used and they are insensitive to the number of atomic levels, provided that number is finite.

The Bloch picture shows clearly that for two-level atoms with or without the rotating-wave approximation, but with spatially independent coupling constants, the problem reduces to that of a single photon mode in the rotating wave approximation. Furthermore, as shown in the Appendix, expectation values of intensive observables can be calculated using the mean fields given by the steepest-descent points of the coherent-state integrals. In this context we note a similarity to those approximations in laser theory in which the photon field is treated semiclassical-

ly, and in which the atoms are treated as independent, but fully quantum-mechanical entities.^{9,10}

Section III explores the question of stability of the more fundamental Hamiltonian in which there are an infinite number of photon modes. Leaving aside the infrared problem, we have established some necessary and some sufficient conditions for the thermodynamic stability. Our methods are not powerful enough, however, to prove the existence of the thermodynamic limit.

In Sec. IV, where Hamiltonians similar to those of Sec. III but with a finite number of photon modes are discussed, the existence of the thermodynamic limit can be established. The main result of Sec. IV is that the inclusion of translational degrees of freedom generally does not destroy the kind of phase transition possessed by the models of Sec. II.

ACKNOWLEDGMENT

The authors would like to thank the I.H.E.S. for its generous hospitality.

APPENDIX: SOME TECHNICAL CONSIDERATIONS AND CALCULATION OF EXPECTATION VALUES

To avoid unnecessary complications we shall consider a one-mode Hamiltonian

$$\begin{aligned} H &= H_0 + R, \\ H_0 &= a^*a, \\ R &= A + a^*B + aB^*, \end{aligned} \tag{A1}$$

where the Hilbert space is $\mathcal{K} = \mathcal{F} \otimes \mathcal{H}$, \mathcal{F} is the Fock space of the boson mode a^* , \mathcal{H} is a finite dimensional Hilbert space of dimension d (the spin space) and A and B are operators on \mathcal{H} with A Hermitian.

Clearly R is a Kato perturbation¹¹ of H_0 , which implies that H is self-adjoint on $D(H_0)$, the domain of H_0 ; H is bounded from below and has purely discrete spectrum with finite multiplicity because H_0 has these properties. Therefore $\rho = e^{-\beta H_0}$ is bounded. To prove that ρ is trace class, write $\rho = \exp[-\frac{1}{2}\beta H_0 - \beta(\frac{1}{2}H_0 + R)]$. By the Golden-Thompson inequality,¹²

$$\text{Tr} \rho \leq \text{Tr} e^{-\beta H_0/2} e^{-\beta(H_0/2 + R)}. \tag{A2}$$

The first factor in (A2) is trace class and the second is bounded.

Let P_n be the projector onto the states with $\leq n$ photons, so that $P_n \rightarrow I$ strongly. Let $H_n = P_n H P_n$ and consider

$$Z_n \equiv \text{Tr} P_n e^{-\beta H_n}, \tag{A3}$$

which is a finite dimensional trace. We note that $D = \bigcup_n P_n \mathcal{K}$ is a common core for all H_n and H and,

for any $\psi \in D$, $P_n H P_n \psi = \psi$ for n sufficiently large. In fact $P_n Q P_n \psi = \psi$ for n sufficiently large when Q is any polynomial in a , with coefficients which are operators on \mathcal{K} . Hence $H_n \rightarrow H$ in the strong resolvent sense.¹³ Let $\{\psi_i, h_i\}_{i=1}^\infty$ (resp. $\{\psi_i^n, h_i^n\}_{i=1}^\infty$) be the eigenvectors and eigenvalues of H (resp. $H_n|P_n\mathcal{K}$) arranged in increasing order. We can use the ψ_i^n as trial vectors for H and conclude from the mini-max principle that $h_i^n \geq h_i$ for $1 \leq i \leq nd$. Using this fact, together with the strong convergence¹³ of the eigenprojections $E_n(a, b) \rightarrow E(a, b)$ for every interval (a, b) with $a, b \in \text{spec}(H)$, one can show that $h_n^i \rightarrow h^i$ and $\psi_n^i \rightarrow \psi^i$ weakly. Hence $Z_n \rightarrow Z$ by the dominated-convergence theorem.

Define the cutoff Glauber states by

$$|\alpha, n\rangle = P_n |\alpha\rangle. \quad (\text{A4})$$

For these states the formulas (2.16)–(2.23) are correct with certain obvious modifications, i.e.,

$$\langle \alpha, n | \beta, n \rangle = K_n(\alpha, \beta) = \exp\left[-\frac{1}{2}|\alpha|^2 - \frac{1}{2}|\beta|^2\right] \times \sum_{m=0}^n (\alpha^* \beta)^m / m!, \quad (\text{A5})$$

$$\text{Tr} |\alpha, n\rangle \langle \beta, n| = K_n(\beta, \alpha), \quad (\text{A6})$$

$$\text{Tr} P_n A P_n = \pi^{-1} \int d\alpha \langle \alpha, n | A | \alpha, n \rangle, \quad (\text{A7})$$

$$\langle \alpha, n | a | \alpha, n \rangle = \alpha K_{n-1}(\alpha, \alpha), \quad (\text{A8})$$

$$\langle \alpha, n | a^* a | \alpha, n \rangle = |\alpha|^2 K_{n-1}(\alpha, \alpha), \quad (\text{A9})$$

$$P_n = \pi^{-1} \int d\alpha |\alpha, n\rangle \langle \alpha, n|, \quad (\text{A10})$$

$$P_n a P_n = \pi^{-1} \int d\alpha \alpha |\alpha, n\rangle \langle \alpha, n|, \quad (\text{A11})$$

$$P_n a^* a P_n = \pi^{-1} \int d\alpha (|\alpha|^2 - 1) |\alpha, n\rangle \langle \alpha, n|, \quad (\text{A12})$$

$$\pi^{-1} \int d\alpha K_n(\beta, \alpha) K_n(\alpha, \gamma) = K_n(\beta, \gamma). \quad (\text{A13})$$

Similar formulas hold for all other polynomials in a^* .

We wish to compute a lower bound to Z_n following Ref. 2. Use (A7) and the fact that $\langle \psi | e^X | \psi \rangle \geq \langle \psi | \psi \rangle \exp(\langle \psi | X | \psi \rangle / \langle \psi | \psi \rangle)$ applied to H_n . Then

$$Z_n \geq \text{Tr}_{\mathcal{K}} \int d\alpha K_n(\alpha, \alpha) \times \exp\left[-|\alpha|^2 K_{n-1}(\alpha, \alpha) / K_n(\alpha, \alpha) + A + [B\alpha^* K_{n-1}(\alpha, \alpha) / K_n(\alpha, \alpha) + \text{H.c.}]\right]. \quad (\text{A14})$$

First we take $\text{Tr}_{\mathcal{K}}$, and then let $n \rightarrow \infty$. By the dominated-convergence theorem, the left side of (2.24) is proved. It is equally clear that the same strategy proves the lower bounds (2.3) and (2.4) obtained from the Bloch state representa-

tion.

It is somewhat more difficult to prove the upper bounds (2.24) or (2.3). According to the method of Ref. 2 one defines

$$Z_n^m = \text{Tr} P_n (1 - \beta H_n / m)^m \quad (\text{A15})$$

and, since the trace is finite dimensional, $\lim_{m \rightarrow \infty} Z_n^m = Z_n$. The difficulty is that if one applies (A5)–(A13) directly to (A15) the dominated convergence theorem will not be clearly valid. Instead, for all $\epsilon \geq 0$ we define $H_n(\epsilon)$ as follows. For each photon operator that appears in H_n , such as $a^* a$ or a^* , one introduces a convergence factor $e^{(-\epsilon |a|^2)}$ in the integrands of (A11) and (A12). Using $H_n(\epsilon)$, one defines $Z_n(\epsilon)$ and $Z_n^m(\epsilon)$. Then $\lim_{\epsilon \downarrow 0} Z_n(\epsilon) = Z_n$ and $\lim_{m \rightarrow \infty} Z_n^m(\epsilon) = Z_n(\epsilon)$. Using the Golden-Thompson inequality, as shown in Ref. 2, one has

$$Z_n^m(\epsilon) \leq \pi^{-1} \int d\alpha K_n(\alpha, \alpha) \text{Tr}_{\mathcal{K}} \{1 - \beta \hat{H}(\alpha, \epsilon)\}^m, \quad (\text{A16})$$

where $\hat{H}(\alpha, \epsilon)$ is defined by the replacements $a^* a \rightarrow (|\alpha|^2 - 1)e^{(-\epsilon |a|^2)}$, etc. Owing to the convergence factor and to the Gaussian decrease of $K_n(\alpha, \alpha)$ [cf. (A5)] one can use dominated convergence to assert that

$$Z_n(\epsilon) \leq \pi^{-1} \int d\alpha K_n(\alpha, \alpha) \text{Tr}_{\mathcal{K}} e^{[-\beta \hat{H}(\alpha, \epsilon)]}. \quad (\text{A17})$$

Again, we can let $\epsilon \downarrow 0$ and use dominated convergence to replace $Z_n(\epsilon)$ by Z_n and $\hat{H}(\alpha, \epsilon)$ by $\hat{H}(\alpha, 0)$ in (A19). Now, because \hat{H} has a term $|\alpha|^2$, we can let $n \rightarrow \infty$ and use dominated convergence to obtain the right-hand side of (2.24). For the Bloch state upper bound, (2.3), the same strategy works except that it is not necessary to introduce the convergence factor, i.e., one can deal directly with Z_n .

Having proved that the bounds on the partition function used in the main text can be justified for the unbounded photon operators, we turn now to the problem of evaluating expectation values of intensive observables. This latter is the algebra generated by $a^*/N^{1/2}$ and by the atomic operators S_N^i/N . In particular we consider an operator Θ which is a monomial in the $a^*/N^{1/2}$ times an operator on \mathcal{K} . Defining

$$\langle \Theta \rangle = \text{Tr} \Theta e^{-\beta H} / \text{Tr} e^{-\beta H}, \quad (\text{A18})$$

we want to show that as $N \rightarrow \infty$, $\langle \Theta \rangle$ can be evaluated by simply replacing each factor in Θ by its value at the absolute maximum of the integrands of (2.11), (2.25), etc., and then summing over those points. [Note that in the Dicke model with the rotating-wave approximation there is a conservation law which leads to a gauge invariance. The

stationary phase "points" are, in fact, curves. After integrating over those curves, one finds, for example, that $N^{-1}\langle a^*a \rangle \neq 0$ but $N^{-1}\langle a^2 \rangle = 0$, as expected (cf. Ref. 1)].

We shall need two lemmas whose use will become clear later. First we give a different proof and a slightly generalized version of Griffiths's lemma.¹⁴

Lemma 1 (Griffiths). Let $\{g_n(x)\}$ be a sequence of convex functions on $x \in (a, b) \equiv I \subset \mathbb{R}$ with a pointwise limit $g(x)$, which, of course, is convex. Let $G_n^+(x)$ [resp. $G_n^-(x)$] be the right (resp. left) derivatives of $g_n(x)$, and similarly for $G^+(x)$, $G^-(x)$. Then, for all $x \in I$,

$$\limsup_{n \rightarrow \infty} G_n^+(x) \leq G^+(x), \quad (\text{A19})$$

$$\liminf_{n \rightarrow \infty} G_n^-(x) \geq G^-(x).$$

In particular, if all the $g_n(x)$ and $g(x)$ are differentiable at some point $x \in I$, then

$$\lim_{n \rightarrow \infty} dg_n(x)/dx = dg(x)/dx. \quad (\text{A20})$$

Proof. Fix $x \in I$. For $y > 0$ and $x \pm y \in I$,

$$g_n(x+y) \geq g_n(x) + yG_n^+(x),$$

$$g_n(x-y) \geq g_n(x) - yG_n^-(x).$$

Fix y and take the limit $n \rightarrow \infty$. Then

$$\limsup_{n \rightarrow \infty} G_n^+(x) \leq y^{-1}[g(x+y) - g(x)]$$

and similarly for $\liminf_{n \rightarrow \infty} G_n^-(x)$. Now let $y \downarrow 0$. Q.E.D.

In the following, Lemma 2, we consider a sequence of "partition functions" which, quite generally, we may write as

$$Z_n(\beta) = \int e^{-\beta n e} d\mu_n(e), \quad (\text{A21})$$

where $\{\mu_n\}$ is some sequence of nonnegative measure on \mathbb{R} . We assume that all $Z_n(\beta) < \infty$ for β in some open interval $I = (a, b)$. Define

$$g_n(\beta) = n^{-1} \ln Z_n(\beta). \quad (\text{A22})$$

Obviously, $g_n(\beta)$ is convex for $\beta \in I$. We also define the moments of the energy (per particle) as

$$E_n^k(\beta) = \int e^k e^{-\beta n e} d\mu_n(e) / Z_n(\beta), \quad (\text{A23})$$

where $k=0, 1, 2, \dots$. It is easy to prove (by dominated convergence, or otherwise) that for $\beta \in I$, $E_n^k(\beta)$ exists and

$$E_n^k(\beta) = (-n)^{-k} Z_n(\beta)^{-1} d^k Z_n(\beta) / d\beta^k. \quad (\text{A24})$$

Lemma 2. Let the sequence $\{\mu_n, Z_n(\beta), g_n(\beta)\}$ be defined as above and assume that

$$\lim_{n \rightarrow \infty} g_n(\beta) = g(\beta) \quad (\text{A25})$$

exists. Then, for every β at which $g(\beta)$ is differentiable,

$$\lim_{n \rightarrow \infty} E_n^k(\beta) = [\lim_{n \rightarrow \infty} E_n^1(\beta)]^k = [-dg(\beta)/d\beta]^k. \quad (\text{A26})$$

Proof. For $k=1$, the lemma is the same as Lemma 1. Let J be a compact subinterval of I . From the facts that $Z_n(\beta) < \infty$ for all $\beta \in I$, and the uniform convergence of $g_n(\beta)$ on compacts, which always holds for convex functions, we can say that if $\beta \in J$ then we can find a lower cutoff e_0 in the integral (A23) such that the error in $g_n(\beta)$ and in $E_n^k(\beta)$ is $O(e^{-cn})$ for all k less than some fixed K , and where $c > 0$ is a constant independent of n and β . By adding a (trivial) constant to e we may assume, without loss of generality, that $e_0 = 1$, i.e., $\mu((-\infty, 1)) = 0$. Then $E_n^k(\beta) > 0$ and $e^k d\mu_n(e)$ is a non-negative measure for all k . Consider $k=2$. Replace $d\mu_n(e)$ by $e d\mu_n(e)$ and thereby define $Z_n^1(\beta)$ and $g_n^1(\beta)$ as in (A23) and (A24). Then $dg_n^1(\beta)/d\beta = -E_n^2(\beta)/E_n^1(\beta)$. Now $\exp[n(g_n^1(\beta) - g_n(\beta))] = Z_n^1(\beta)/Z_n(\beta)$, so $h_n(\beta) \equiv g_n^1(\beta) - g_n(\beta) \rightarrow 0$. Hence, $g_n^1(\beta) = h_n(\beta) + g_n(\beta)$ is a convex function having the limit $g(\beta)$, and we can apply Lemma 1 to it on $\text{int}(J)$. This proves the $k=2$ case. Obviously, the argument can be extended inductively, i.e., one writes $E_n^k(\beta) = \prod_{j=1}^k [E_n^{k-j+1}(\beta)/E_n^{k-j}(\beta)]$. Q.E.D.

Now we return to the monomial Θ whose expectation value we wish to compute. If Θ is not self-adjoint then consider Θ to be replaced by $\Theta + \Theta^*$. Clearly there is some integer $k \geq 1$ such that H^k dominates Θ , i.e., $|\Theta| \leq \frac{1}{2}H^k + bI$ for some positive constant b . With $\beta > 0$ fixed, let $e_n = \lim_{N \rightarrow \infty} N^{-k} \langle H^k \rangle$. For each $\lambda \geq 0$ define $H(\lambda) \equiv H + \lambda N \Theta + \lambda W$, with $W = N^{-k+1}H^k - e_{k-1}H$. Obviously, the preceding analysis for Z applies equally well to $Z(\lambda) \equiv \text{Tr} e^{-\beta H(\lambda)}$, i.e., one can obtain upper and lower bounds to $Z(\lambda)$ in terms of classical integrals. Naturally, for the monomials in Θ and H^k one has to find the obvious generalizations of (2.19)–(2.23), but this can easily be done; in particular the monomial $(a^*)^p a^q$ has $(\alpha^*)^p \alpha^q$ as its leading term in (2.19)–(2.23).

The Peierls-Bogoliubov inequality

$$Z(\lambda) \geq Z(0) e^{-\beta \lambda \langle W \rangle}, \quad (\text{A27})$$

can easily be proved to hold here. Then, with $g(\beta, N, \lambda) \equiv N^{-1} \ln Z(\lambda)$ and $\lambda > 0$,

$$\beta \langle \Theta \rangle \geq \lambda^{-1} [g(\beta, N, 0) - g(\beta, N, \lambda)] - \beta N^{-1} \langle W \rangle. \quad (\text{A28})$$

Take the limit $N \rightarrow \infty$. In those cases (Secs. II and IV) in which our upper and lower bounds agree, they will also agree when $\lambda > 0$. Thus, $\lim_{N \rightarrow \infty} g(\beta, N, \lambda) = g(\beta, \lambda)$ exists. Also, $\lim_{N \rightarrow \infty} N^{-1} \langle W \rangle = 0$ by Lemma 2. Thus, for all $\lambda > 0$

$$\liminf_{N \rightarrow \infty} \langle \Theta \rangle \geq \lambda^{-1} [g(\beta, \lambda) - g(\beta, 0)]. \quad (\text{A29})$$

In a parallel way, we can define $H'(\lambda) = H - \lambda N \Theta + \lambda W$ and $g'(\beta, N, \lambda)$, and obtain

$$\limsup_{N \rightarrow \infty} \langle \Theta \rangle \leq \lambda^{-1} [g(\beta, 0) - g'(\beta, \lambda)]. \quad (\text{A30})$$

To complete the demonstration one has to show that as $\lambda \downarrow 0$ the right-hand sides of (A29) and (A30) have a common limit and that this is the classical value of $\langle \Theta \rangle$ at the steepest-descent points for $\lambda = 0$. When these points are isolated, as they are in the absence of the rotating-wave approximation, it is easy to see that the above is true; as $\lambda \downarrow 0$ some points approach the $\lambda = 0$ points while others go off to infinity. The latter are not maximal, however. In the rotating-wave approximation, the $\lambda = 0$ points are in fact curves $|\alpha| = \text{const}$. In this case, (A29) and (A30) will agree for those operators such as a^*a/N or $aS^+/N^{3/2}$ which are invariant under the gauge group. For other operators, such as a^2 , the two limits will not agree because as $\lambda \downarrow 0$ the steepest-descent curves will be approached as two different

points. However, in such cases it is easy to see directly that $\langle a^2 \rangle = 0$, and this value agrees with what one would get if one integrated over the entire curve.

This completes our discussion of the Hamiltonian (A1) and clearly no difficulty is encountered in extending it to the multimode case. However, some remarks are needed for the Hamiltonians considered in Secs. III and IV because the unbounded operators T and/or U are introduced. The additional Hilbert space, \mathcal{G} , is generally $L^2(V^N)$, where V is the box, but if U has a hard core it is $L^2(V^N - \text{hard-core region})$. In either case, it suffices to say that in addition to the P_n photon projection operators one can also introduce a strongly convergent sequence of projections $\{Q_n\}$ on \mathcal{G} such that $D = \bigcup_n Q_n \mathcal{G}$ is a common core for H and all $Q_n H Q_n$ and such that $(Q_n H Q_n - H)\varphi \rightarrow 0$ strongly for all $\varphi \in D$.

The remarks in this appendix are admittedly sketchy in places, but the interested reader can easily fill in the details required to analyze any particular model of the class we have considered in the main text.

*On leave from the Department of Mathematics, MIT, Cambridge, Mass. 02139, U. S. A. Work partially supported by U. S. National Science Foundation under Grant No. GP-31674X and by a Guggenheim Memorial Foundation Fellowship.

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