still about 15 Å; so one might have expected Boltzmann's assumptions to be more nearly justified than in many applications to plasmas and solids. Our results suggest that, instead of seeking to characterize the *physical systems* for which the Boltzmann transport equation is valid, it may be more appropriate to seek, for a given physical system, to characterize the *initial states* for which it is valid. The analysis given here, of course, is very far from answering this question.

*Work supported by the Air Force Office of Scientific Research, Contract No. F44620-60-C-0121.

[1]E. T. Jaynes, in *Statistical Physics*, edited by K. W. Ford (Benjamin, New York, 1963), Vol. 3, Chap. 4.

[2]This assumption appears to be one of the "fundamental" propositions of statistical mechanics; i.e., one which has never been proved or doubted; and we do neither here. We have, of course, the well-known fact that the Maxwellian distribution with a particular partitioning of total energy between kinetic and potential, corresponds to an overwhelmingly greater phase volume than any other disposition of the energy; this makes it highly plausible, but does not prove, that a physical system will go to this equilibrium condition. In addition, we have excellent experimental evidence that the assumption is correct for all systems; otherwise it is very hard to see how the propositions of elementary thermodynamics (existence of a definite, reproducible equation of state, heat capacity, etc.) could be valid.

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PHYSICAL REVIEW A VOLUME 4, NUMBER 2 AUGUST 1971

Theory of Superfluidity

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The generator-coordinate method, well known in nuclear physics, is employed to derive the Bogoliubov theory of superfluidity of a boson system. The treatment is fully number conserving. Analogies to a *normal* Fermi system (especially to the random-phase approximation for that system) are pointed out, and some further insight into the nature of superfluidity is gained. Possible generalizations are mentioned.

I. INTRODUCTION

The generator-coordinate method[1–4] is well known as a powerful means of treating nuclear systems, and is especially useful for describing collective properties of nuclei. One of its virtues is a simultaneous treatment of both the ground state and the low-lying states of the system. One of its applications was an alternative derivation, by Jancovici and Schiff, [5] of the random-phase approximation (RPA) for a normal Fermi system. Extensions of the Jancovici-Schiff treatment to a derivation of a higher RPA[6] as well as to a pairing Fermi system[7] have been suggested. There is nothing in the method which would limit its usefulness to the realm of nuclear physics only. In fact, it may find applications in various other many-body systems. The purpose of the present paper is to call attention to the applicability of the method to a many-boson system and, in particular, to show that a fully number-conserving application of the method, in a form closely analogous to the Jancovici-Schiff derivation of the RPA, yields a number-conserving version of the Bogoliubov theory of superfluidity.[8–11] The relation between the Bogoliubov transformation in a boson system and the *summation* of the RPA *diagrams* has been known for a long time.[9,12] However, up to now it has been derived within the context of a particle number nonconserving formalism, which keeps only the *average* number of particles fixed. It is interesting to note that the Jancovici-Schiff derivation of the RPA for a Fermi system does *not* yield the pairing effects. The BCS and Bogoliubov-Valatin results in the theory of superconductivity are derived only at the expense of introducing states which do not conserve the number of particles. Usually, the Bogoliubov-Valatin quasiparticles in the theory of superconductivity are considered the analogs of the quasiparticles representing the low-lying states of a superfluid boson system. However, the present formalism implies that the latter states are rather the analogs of the RPA collective particle-hole modes of a *normal* Fermi system. Although we will not carry it out in the present publication, it is tempting to include number nonconservation in the present formalism, in the manner used in Ref. 7 for the discussion of the so-called "pairing vibrations"

of a Fermi system.

In the rest of this section, we review very briefly the generator-coordinate method. In Sec. II we shall see how a particular approximation scheme yields, in a number-conserving formalism, the Bogoliubov theory of superfluidity. Section III discusses the results and indicates some possible generalizations.

In the generator-coordinate method[1] one starts with a known family of N-particle wave functions $|\Phi(\alpha)\rangle$, where α stands for a set of parameters (generator coordinates) $\alpha_1, \alpha_2, \ldots$. An approximate eigenfunction of the system is then generated by taking the linear combination

$$|\psi\rangle = \int f(\alpha)\,|\Phi(\alpha)\rangle\, d\alpha \ . \tag{1.1}$$

The unknown weight function $f(\alpha)$ is to be determined from the variational principle

$$\delta[\langle\psi|H|\psi\rangle / \langle\psi|\psi\rangle] = 0 \ . \tag{1.2}$$

This leads to an integral equation for $f(\alpha)$,

$$\int [H(\alpha,\alpha')/I(\alpha,\alpha') - E] I(\alpha,\alpha') f(\alpha')\, d\alpha' = 0 \ . \tag{1.3}$$

where

$$\begin{aligned} H(\alpha,\alpha') &= \langle\Phi(\alpha)|H|\Phi(\alpha')\rangle \ , \\ I(\alpha,\alpha') &= \langle\Phi(\alpha)|\Phi(\alpha')\rangle . \end{aligned} \tag{1.4}$$

The lowest eigenvalue E of Eq. (1.3) should give an approximation to the ground-state energy and higher eigenvalues will approximate the energies of excited states. Of course, the usefulness and tractability of the method depend to a large extent on the choice of the set $|\Phi(\alpha)\rangle$.

II. GENERATOR-COORDINATE METHOD AND BOGOLIUBOV THEORY OF SUPERFLUIDITY

Consider a system of N spinless bosons enclosed in a cubic box of volume Ω and described by the Hamiltonian

$$H = \sum_{\vec{k}} \frac{\vec{k}^2}{2m} + \tfrac{1}{2}\Omega^{-1} \sum_{\vec{k}\vec{p}\vec{q}} V_{\vec{k}}\, a^\dagger_{\vec{p}+\vec{k}}\, a^\dagger_{\vec{q}-\vec{k}}\, a_{\vec{p}}\, a_{\vec{q}} \ . \tag{2.1}$$

(We assume periodic boundary conditions and take $V_{\vec{k}} = V_{-\vec{k}} = V^*_{\vec{k}}$.) The ground state of the noninteracting system is $|\Phi_0\rangle$,

$$|\Phi_0\rangle = (N!)^{-1/2} (a_0^\dagger)^N |0\rangle \ , \tag{2.2}$$

where $|0\rangle$ is the vacuum state. The most general state which is not orthogonal to $|\Phi_0\rangle$ may be written (up to normalization) in the form

$$\exp\Big(\Omega^{-1/2}\sum_{\vec{k}\neq 0} z_{\vec{k}}\, a^\dagger_{\vec{k}}\, a_0 + \tfrac{1}{2}\Omega^{-1}\sum_{\vec{k},\vec{p}\neq 0} z_{\vec{k},\vec{p}}\, a^\dagger_{\vec{k}}\, a^\dagger_{\vec{p}}\, a_0^2 + \cdots\Big)|\Phi_0\rangle \ .$$

In order to derive the Bogoliubov results in the theory of superfluidity, we take the generator coordinates to be the (simplest choice) complex amplitudes $z_{\vec{k}}$, and take $|\Phi(\alpha)\rangle$ to be the state

$$|\Phi(z)\rangle \equiv \exp\Big(N^{-1/2}\sum_{\vec{k}\neq 0} z_{\vec{k}}\, a^\dagger_{\vec{k}}\, a_0\Big)|\Phi_0\rangle \ . \tag{2.3}$$

[Note that in Eq. (2.3) we chose the coefficient of the sum in the exponent to be $N^{-1/2}$ instead of $\Omega^{-1/2}$. This makes the expressions in the intermediate stages of the calculation somewhat simpler and does not affect any of the results.] As the trial wave function for the generator-coordinate method we take

$$|\psi\rangle = \int f(z)\,|\Phi(z)\rangle\, dz \ , \tag{2.4}$$

where dz means $d(\mathrm{Re}z)\, d(\mathrm{Im}z)$. Notice that the $z_{\vec{k}}$ here are the exact analogs of the complex particle-hole amplitudes z_{mi} of the RPA.[5] The variational principle yields the following integral equation for $f(z)$

$$\int [H(z,z')/I(z,z') - E] I(z,z') f(z')\, dz' = 0 \ , \tag{2.5}$$

where

$$H(z,z') = \langle\Phi(z)|H|\Phi(z')\rangle \ , \quad I(z,z') = \langle\Phi(z)|\Phi(z')\rangle \ . \tag{2.6}$$

Assume now that even in the presence of the interaction the state $|\Phi_0\rangle$ is not too bad an approximation to the real ground state (this assumption is of course, related to the usual assumption of macroscopic occupancy of the $\vec{k}=0$ mode at $T < T_\lambda$ even in the presence of interactions), and that most of the contribution to the real ground state comes from the neighborhood of $z=0$. This would mean that the function $f(z)$ of Eq. (2.4) should be peaked around the point $z=0$.[13] These assumptions are essentially equivalent to assuming z, z' to be small, and that we may expand both $I(z,z')$ and $H(z,z')/I(z,z')$ in Eq. (2.5) up to second order in z, z', neglecting higher-order terms.

A simple calculation yields that up to second order in z, z', $I(z,z')$ may be written in the form

$$I(z,z') = \exp\Big(\sum_{\vec{k}\neq 0} z^*_{\vec{k}}\, z'_{\vec{k}}\Big) \ , \tag{2.7}$$

while, to the same order, $H(z,z')/I(z,z')$ assumes the form

$$\frac{H(z,z')}{I(z,z')} = E_0 + \sum_{\vec{k}\neq 0}\Big(\frac{\vec{k}^2}{2m} + (N-1)\Omega^{-1} V_{\vec{k}}\Big) z^*_{\vec{k}}\, z'_{\vec{k}} + \tfrac{1}{2}(N-1)\Omega^{-1}\sum_{\vec{k}\neq 0} V_{\vec{k}}\,(z^*_{\vec{k}}\, z^*_{-\vec{k}} + z'_{\vec{k}}\, z'_{-\vec{k}}) \ , \tag{2.8}$$

where

$$E_0 = \langle\Phi_0|H|\Phi_0\rangle = \tfrac{1}{2}\Omega^{-1} V_0 (N^2 - N) \ .$$

Therefore, the integral equation (2.5) reads in our approximation

$$\int\Big[\sum_{\vec{k}\neq 0}\Big(\frac{\vec{k}^2}{2m}+(N-1)\Omega^{-1}V_{\vec{k}}\Big)z_{\vec{k}}^{*}z_{\vec{k}}' + \tfrac{1}{2}(N-1)\Omega^{-1}\sum_{\vec{k}\neq 0}V_{\vec{k}}(z_{\vec{k}}^{*}z_{-\vec{k}}^{*}+z_{\vec{k}}'z_{-\vec{k}}')-(E-E_0)\Big] \times\exp\Big(\sum_{\vec{k}\neq 0}z_{\vec{k}}^{*}z_{\vec{k}}'\Big)f(z')dz'=0\ . \quad (2.9)$$

The integral equation (2.9) is equivalent to an equation for coupled harmonic oscillators.[5] Define a function G,

$$G(z^{*})=\int\exp\Big(\sum_{\vec{k}\neq 0}z_{\vec{k}}^{*}z_{\vec{k}}'\Big)f(z')dz'\ . \quad (2.10)$$

Obviously,

$$\frac{\delta G}{\delta z_{\vec{k}}^{*}}=\int z_{\vec{k}}'\exp\Big(\sum_{\vec{k}\neq 0}z_{\vec{k}}^{*}z_{\vec{k}}'\Big)f(z')dz'\ . \quad (2.11)$$

By means of this relation, the integral equation (2.9) for f is transformed into the partial differential equation for G:

$$\Big[\sum_{\vec{k}\neq 0}\Big(\frac{\vec{k}^2}{2m}+(N-1)\Omega^{-1}V_{\vec{k}}\Big)z_{\vec{k}}^{*}\frac{\delta}{\delta z_{\vec{k}}^{*}} + \tfrac{1}{2}(N-1)\Omega^{-1}\sum_{\vec{k}\neq 0}V_{\vec{k}}\Big(z_{\vec{k}}^{*}z_{-\vec{k}}^{*}+\frac{\delta}{\delta z_{\vec{k}}^{*}}\frac{\delta}{\delta z_{-\vec{k}}^{*}}\Big) -(E-E_0)\Big]G(z^{*})=0\ . \quad (2.12)$$

Equation (2.12) is the Schrödinger equation for coupled harmonic oscillators in the Bargmann representation.[5,14,15] To diagonalize it, define the boson operator $B_{\vec{k}}^{\dagger}$,

$$B_{\vec{k}}^{\dagger}=u_{\vec{k}}z_{\vec{k}}^{*}+v_{\vec{k}}\delta/\delta z_{-\vec{k}}^{*}\ . \quad (2.13)$$

It is easy to see that Eq. (2.12) reads, in terms of the new operators,

$$\Big[\sum_{\vec{k}}\omega_{\vec{k}}(B_{\vec{k}}^{\dagger}B_{\vec{k}}+\tfrac{1}{2})-\frac{1}{2}\Big(\frac{\vec{k}^2}{2m}+(N-1)\Omega^{-1}V_{\vec{k}}\Big) -(E-E_0)\Big]G=0\ , \quad (2.14)$$

where the excitation energy $\omega_{\vec{k}}$ is given by

$$\omega_{\vec{k}}=[(N-1)\Omega^{-1}V_{\vec{k}}\,\vec{k}^2/m+\vec{k}^4/4m^2]^{1/2} \quad (2.15)$$

and the coefficients $u_{\vec{k}}$, $v_{\vec{k}}$ are given by

$$u_{\vec{k}}=(1-X_{\vec{k}}^2)^{-1/2}\ ,\quad v_{\vec{k}}=X_{\vec{k}}(1-X_{\vec{k}}^2)^{-1/2}\ , \quad (2.16)$$

where

$$X_{\vec{k}}=\frac{(N-1)\Omega^{-1}V_{\vec{k}}}{\vec{k}^2/2m+(N-1)\Omega^{-1}V_{\vec{k}}+\omega_{\vec{k}}}\ . \quad (2.17)$$

From Eqs. (2.14) and (2.15), it is obvious that the energy spectrum obtained is the same as that of Bogoliubov.[8,9] We can also obtain the corresponding wave functions.[5] The function $G_0(z)$ which corresponds to the ground state is the solution of

$$B_{\vec{k}}G_0(z^{*})=0\ , \quad (2.18)$$

and the excited states are reached by the application of the operators $B_{\vec{k}}^{\dagger}$ to G_0. Equation (2.18) is easily seen to yield (up to normalization)

$$G_0(z^{*})=\exp\Big(-\tfrac{1}{2}\sum_{\vec{k}\neq 0}X_{\vec{k}}z_{\vec{k}}^{*}z_{-\vec{k}}^{*}\Big)\ . \quad (2.19)$$

Finally, once we know $G(z^{*})$, we are able to find $f(z)$ and $|\psi\rangle$.[5] One obtains

$$|\psi\rangle=G(N^{-1/2}a_{\vec{k}}^{\dagger}a_0)|\Phi_0\rangle\ , \quad (2.20)$$

where $G(N^{-1/2}a_{\vec{k}}^{\dagger}a_0)$ is the operator obtained through the replacement in $G(z^{*})$ of each $z_{\vec{k}}^{*}$ by the operator $N^{-1/2}a_{\vec{k}}^{\dagger}a_0$. In particular, the ground state is given in our approximation by

$$|\psi\rangle=\exp\Big(-\tfrac{1}{2}N^{-1}\sum_{\vec{k}\neq 0}X_{\vec{k}}a_{\vec{k}}^{\dagger}a_{-\vec{k}}^{\dagger}a_0^{2}\Big)|\Phi_0\rangle\ . \quad (2.21)$$

III. DISCUSSION

We have already pointed out the analogy between the present formulation of superfluidity and the RPA for a normal Fermi system. Let us elaborate somewhat further on this analogy, especially in relation to a pairing (superconductive) Fermi system. Superconductivity in a fermion system is brought about by the interaction of two *particles* (or two *holes*)[16] of equal and opposite momenta, leading to the formation of Cooper pairs. On the other hand, in the present formulation (at least) superfluidity is seen to be caused by[17] a "particle"-"hole" interaction. Here, "particles" have momentum $\vec{k}\neq 0$ and "holes" are created in the condensate (the macroscopically occupied $\vec{k}=0$ mode) by taking particles out of it, in much the same manner that holes are created in a Fermi system, with the condensate playing here the role of a Fermi sea. This "particle"-"hole" interaction is intimately connected with number conservation, since without number conservation there would not be "holes" in the condensate.[18] Furthermore, although the wave function $|\psi\rangle$ in Eq. (2.21) looks to be formally of the "pairing" type, this form is necessitated by *momentum conservation*, and actually $|\psi\rangle$ is of the form of the RPA ground state of a normal Fermi system.

Let us note another amusing similarity to a Fermi system. Had we assumed the operator $N^{-1/2}a_{\vec{k}}^{\dagger}a_0$ to be a boson creation operator,[19] Eq. (2.7) would have been exact. This is of course the analog of the quasiboson approximation in a Fermi system, and it raises the possibility of a *boson expansion* in a boson system.[20,21]

Finally, let us mention briefly some possible generalizations of our treatment. These are related to generalizing $|\Phi_0\rangle$ and the set $|\Phi(z)\rangle$ to include more correlations, for example in the form suggested in Ref. 6 for deriving a higher RPA.

Another possibility is to use the generator-coordinate method to treat the interaction between various collective modes as suggested in Refs. 22 and 6. Last but not least, the Feynman[23] and Feynman-Cohen[24] approach to liquid helium is easily seen to fall within the formalism of the generator-coordinate method if one identifies the generator coordinates with the coordinates of the atoms in the liquid.

ACKNOWLEDGMENT

It is a pleasure to thank Professor F. B. Malik for his comments on the manuscript.

*Work supported in part by the U. S. Army Research Office, Durham, N. C.

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PHYSICAL REVIEW A VOLUME 4, NUMBER 2 AUGUST 1971

Nonlinear Optical Polarization and Relaxation Phenomena in Dilute Gases

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The Kubo-Zwanzig-Fano relaxation method is extended to the problem of a homogeneous dilute system interacting with arbitrarily strong fields composed of several monochromatic modes, neglecting breakdown and spatial correlations. The Von Neumann equation for the density matrix is solved for classical fields, using a procedure in which the harmonics numbers are treated as the classical analog (in Liouville space) of the quantum fields occupation-number representation. This solution is adapted to the case where memory effects during the absorption or emission of a photon can be neglected. Two examples illustrate the method, which is generally applicable in such problems as saturation, double resonance, and frequency mixing involving the nonlinear response of independent molecules.

I. INTRODUCTION

The understanding of relaxation phenomena in resonance spectra of gases and other dilute systems in the linear-response approximation has been improved considerably by the introduction of oper-