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Equilibrium Superradiance in a Bose Gas

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A model of free ^{4}He atoms interacting with radiation exhibits an equilibrium phase transition in which the atomic ground-state Bose condensation is coupled to condensations of virtual photons and virtually excited atoms of the same macroscopic wavelength. The condensed phase has a twofold polarization degeneracy. It is suggested that this might furnish a mechanism for a discrete symmetry-related phase degeneracy of superfluid liquid ^{4}He required to explain the λ transition according to Tisza's generalized Gibbsian thermodynamics. A more realistic model would require inclusion of repulsive interactions.

KEY WORDS: Equilibrium superradiance; Bose condensation; liquid helium-4.

Lambda-type specific heat anomalies (logarithmic specific heat singularities) are associated with order–disorder transitions characterized by the existence at temperatures $T < T_\lambda$ of degenerate phases differing only in some discrete symmetry parameter. Tisza has presented[1] a generalized form of Gibbsian thermodynamics which implies that a λ transition can *only* arise from the onset of such a discrete phase degeneracy. He concluded that such a degeneracy must be present in the superfluid phase of liquid ^{4}He. We wish to propose a possible mechanism for such a degeneracy.

We start from the observation that helium atoms possess degenerate excited states. Might this induce a related degeneracy of the many-atom ground state? The atomic excited states are usually not included explicitly in theories of liquid ^{4}He because their excitation energies are so high ($\sim 10^5$ K). Note, however, that *virtually* excited atomic states are responsible for the fact that the system is a liquid, since the attractive van der Waals

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interaction (London dispersion force) results from correlated virtual dipole interactions. Might some nonzero fraction of the virtual atom–radiation interactions in superfluid liquid ^{4}He take place in a coherent fashion, requiring an explicit treatment of the corresponding photon mode?

There exist models of "equilibrium superradiance," e.g., the Dicke model,[2–5] which exhibit phase transitions in which the low-temperature phase contains a macroscopic number of photons in a single mode (Bose condensation of photons). In equilibrium these condensed photons are virtual and do not result in radiation in excess of thermal radiation. Nevertheless, they share with a real lasing mode the properties of coherence and polarization, and lead to a coupling of polarization of virtually excited atomic states through their coupling with the polarized photon mode.

Might such a phenomenon be a candidate for a discrete, symmetry-related degeneracy of superfluid liquid ^{4}He as required by the Tisza theory? Such a possibility is suggested by the absence of Doppler broadening of virtual photons emitted and absorbed by atoms in the zero-momentum ground-state condensate; this might favor the formation of an associated virtual photon condensate as well as a condensate of virtually excited atoms.

The Hamiltonian of a system of ^{4}He atoms interacting with each other and with the quantized radiation field can be derived from first principles by application of an appropriate unitary transformation[6,7] to the Hamiltonian of a system of nuclei, electrons, and radiation in Coulomb gauge. The resultant Hamiltonian is an infinite series representing all physically possible processes. The atom–radiation part starts with terms representing emission, absorption, and scattering of radiation by single atoms as, e.g., exhibited previously by Nakajima,[8] but there are also terms representing photo-ionization, simultaneous interaction of two or more colliding atoms with radiation, etc. The derivation and discussion will be given elsewhere. For the present, we shall exhibit only the terms crucial for the effects we wish to discuss, and hence adopt the model Hamiltonian

$$
\begin{aligned}
H = \sum_{\mathbf{k}} \left[\frac{\hbar^2 k^2}{2m} a_{\mathbf{k}}{}^{\dagger} a_{\mathbf{k}} + \sum_{\lambda} \left(\hbar\omega_0 + \frac{\hbar^2 k^2}{2m} \right) a_{\mathbf{k}\lambda}^{\dagger} a_{\mathbf{k}\lambda} + \sum_{\lambda} \hbar c k b_{\mathbf{k}\lambda}^{\dagger} b_{\mathbf{k}\lambda} \right] \\
+ \hbar\omega_0 \sum_{\mathbf{k}\lambda}{}' \left(\frac{\pi\alpha\omega_0}{ck\Omega} \right)^{1/2} (a_{\mathbf{k}\lambda}^{\dagger} b_{\mathbf{k}\lambda} a_0 + a_0 b_{\mathbf{k}\lambda}^{\dagger} a_{\mathbf{k}\lambda}) \\
+ \sum_{\mathbf{k}\lambda}{}' \frac{2\pi\alpha' \hbar {\omega_0}^2}{ck\Omega} b_{\mathbf{k}\lambda}^{\dagger} b_{\mathbf{k}\lambda} \left(a_0{}^{\dagger} a_0 + \sum_{\lambda'} a_{\mathbf{k}\lambda'}^{\dagger} a_{\mathbf{k}\lambda'} \right) \qquad (1)
\end{aligned}
$$

where the allowed $\mathbf{k}$ values are determined by periodic boundary conditions for a macroscopic cubical volume Ω, and $a_{\mathbf{k}}$ refer to unexcited atoms, the $a_{\mathbf{k}\lambda}$ to those excited to the lowest spin-singlet P state with excitation energy $\hbar\omega_0$, and the $b_{\mathbf{k}\lambda}$ refer to photons. The index $\lambda = 1, 2$ labels the two transverse polarization states; only excited atoms with transverse P orbitals are included since the longitudinal ones do not couple to the radiation field. The terms involving the P-state polarizability α arise from the term in the matter–radiation interaction linear in the vector potential A; we have retained only terms representing virtual excitation from and deexcitation back to the $\mathbf{k} = 0$ condensate. The terms proportional to $\alpha' = 2e^2/m_e\omega_0{}^2$ (m_e is the electron mass) are quadratic in A; we have retained only diagonal terms referring to the ground-state condensate and to the same excited states included in the A term. The ^{4}He-atom annihilation and creation operators $a_\mathbf{k}$, $a_\mathbf{k}{}^\dagger$, $a_{\mathbf{k}\lambda}$, and $a_{\mathbf{k}\lambda}^\dagger$ satisfy the usual Bose commutation relations and commute with the photon annihilation and creation operators $b_{\mathbf{k}\lambda}$ and $b_{\mathbf{k}\lambda}^\dagger$, the algebraic complications associated with the internal structure of the ^{4}He atoms having been transferred from the commutation relations to the Hamiltonian by an appropriate unitary transformation.[6,7]

Consider a variational trial ground state coherent with respect to the ground-state condensate and also with respect to possible momentum-$\mathbf{q}$ condensates of ^{4}He atoms and photons:

$$|\psi_0\rangle = \text{const} \times \exp(\sqrt{n}\,\alpha_0 a_0{}^\dagger) \exp\Big(-\sqrt{n} \sum_\lambda \alpha_\lambda a_{\mathbf{q}\lambda}^\dagger\Big) \times \exp\Big(\sqrt{n} \sum_\lambda \beta_\lambda b_{\mathbf{q}\lambda}^\dagger\Big) |0\rangle \tag{2}$$

Here n is the number of ^{4}He atoms and α_0, α_λ, β_λ, and q are the variational parameters. The expectation value of (1) in such a state yields

$$\frac{E_0}{n\hbar\omega_0} = \left(1 + \frac{\hbar\omega_0 z^2}{2mc^2}\right) \sum_\lambda |\alpha_\lambda|^2 + (z + 2\pi\rho\alpha' z^{-1}) \sum_\lambda |\beta_\lambda|^2 - (\pi\rho\alpha)^{1/2} z^{-1/2} \sum_\lambda (\alpha_\lambda{}^* \beta_\lambda \alpha_0 + \alpha_0{}^* \beta_\lambda{}^* \alpha_\lambda) \tag{3}$$

where $\rho = n/\Omega$ = number density of atoms, $z = cq/\omega_0$, and use has been made of the atom-number conservation condition

$$|\alpha_0|^2 + \sum_\lambda |\alpha_\lambda|^2 = 1 \tag{4}$$

The energy expression (3) exhibits the usual high degeneracy associated with the gauge transformation of the first kind $\alpha_0 \to \alpha_0 e^{i\theta}$, $\alpha_\lambda \to \alpha_\lambda e^{i\theta}$, as well as a twofold polarization degeneracy associated with rotations $\alpha_1 \to \alpha_1 \cos\gamma + \alpha_2 \sin\gamma$, $\beta_1 \to \beta_1 \cos\gamma + \beta_2 \sin\gamma$, etc., and a directional degeneracy with respect to the direction of $\mathbf{q}$. This latter degeneracy is, strictly speaking, partially lifted by the discreteness of $\mathbf{k}$ space. Of these three types of degeneracy, the one that seems to us to be a candidate for the discrete symmetry-related degeneracy sought by Tisza[1] is the twofold polarization degeneracy.

Choosing new real, positive variables x_0, x_1, y_1 such that $\alpha_0 = x_0$, $\alpha_1 = x_1$, $\beta_1 = z^{1/2}y_1$, $\alpha_2 = \beta_2 = 0$, one has

$$\frac{E_0}{n\hbar\omega_0} = \left(1 + \frac{\hbar\omega_0 z^2}{2mc^2}\right)x_1^2 + (z^2 + 2\pi\rho\alpha')y_1^2 - 2(\pi\rho\alpha)^{1/2}x_1 y_1(1 - x_1^2)^{1/2} \quad (5)$$

The minimum with respect to z occurs at $z \to 0+$, and minimization of the resultant expression with respect to y_1 after eliminating x_0 via (4) yields

$$y_1 = \frac{(\pi\rho\alpha)^{1/2}}{2\pi\rho\alpha'} x_1(1 - x_1^2)^{1/2} \quad (6)$$

and

$$\frac{E_0}{n\hbar\omega_0} = -\left(\frac{\alpha - 2\alpha'}{2\alpha'}\right)x_1^2 + \left(\frac{\alpha}{2\alpha'}\right)x_1^4 \quad (7)$$

We must now distinguish two cases. If $\alpha < 2\alpha'$, then the minimum occurs at $x_1^2 = 0$, corresponding to the normal Bose condensate solution. On the other hand, if $\alpha > 2\alpha' > 0$, then the minimum is at

$$x_1^2 = \frac{\alpha - 2\alpha'}{2\alpha}, \qquad \frac{E_0}{n\hbar\omega_0} = -\frac{(\alpha - 2\alpha')^2}{8\alpha\alpha'} \quad (8)$$

In order to correctly elucidate the structure of the corresponding condensate, one must recognize that the minimum allowed value of z is not zero, but $cq_{\min}/\omega_0$, where $q_{\min} = 2\pi\Omega^{-1/3}$, since photons of zero wave vector do not exist. One then finds the mean occupation numbers of the condensates in the variational ground state to be

$$\langle a_0{}^\dagger a_0\rangle_0 = nx_0^2 = n\frac{\alpha + 2\alpha'}{2\alpha}, \qquad \langle a_{\mathbf{q}1}^\dagger a_{\mathbf{q}1}\rangle_0 = nx_1^2 = n\frac{\alpha - 2\alpha'}{2\alpha}$$

$$\langle b_{\mathbf{q}1}^\dagger b_{\mathbf{q}1}\rangle_0 = z_{\min} n y_1^2 = n^{2/3}\frac{2\pi c\rho^{1/3}}{\omega_0}\frac{\alpha^2 - 4(\alpha')^2}{16\pi\rho\alpha(\alpha')^2} \quad (9)$$

This may be called a superradiant phase since the coherent virtual photon mode has an occupation $O(n^{2/3})$ far in excess of the normal $O(1)$ mode occupations; however, it is a less extreme condensation than the macroscopic $O(n)$ population of the superradiant mode in the Dicke model.[2–5] One may picture this solution as representing a situation in which a highly occupied virtual photon mode of macroscopic wavelength $\Omega^{1/3}$ interacts with zero-momentum ground-state condensate atoms, leading to virtual excitation of a fraction nx_1^2 of them into an "exciton condensate" mode of the same macroscopic wavelength. It is perhaps more correct to regard this exciton condensate and the photon condensate as together constituting a hybrid condensate of wave vector $\mathbf{q}_{\min}$, in which momentum is passed back and forth between virtually excited ^{4}He atoms and photons; when a virtually excited atom passes its momentum to the photon condensate, it drops back into the atomic ground-state zero-momentum condensate.

The analysis thus far presented breaks down at the exceptional point $\alpha' = 0$; however, a more careful treatment of the limit $z \to 0+$ in that case shows that E_0 and the occupations all remain finite, but with $E_0 = -O(n^{5/3})$, $\langle b_{\mathbf{q}1}^\dagger b_{\mathbf{q}1}\rangle_0 = O(n^2)$, and $\langle a_0{}^\dagger a_0\rangle_0$ and $\langle a_{\mathbf{q}1}^\dagger a_{\mathbf{q}1}\rangle_0$ both equal to $(1/2)n$. This is a highly pathological "hyperradiant" state and will not be discussed further since the case $\alpha' = 0$ is not physically realizable.

Although our results were derived from a variational Ansatz (2), we believe that they represent the exact ground state of the model Hamiltonian (1) in the thermodynamic limit $n \to \infty$, $\Omega \to \infty$, $n/\Omega \to \rho$, $0 < \rho < \infty$.

The analysis is easily extended to nonzero temperature by the Gibbs–Bogoliubov variational principle (see, e.g., Ref. 9). Performing the unitary transformation

$$U^{-1}a_0 U = a_0 + \sqrt{n}\,\alpha_0, \qquad U^{-1}a_{\mathbf{q}\lambda}U = a_{\mathbf{q}\lambda} + \sqrt{n}\,\alpha_\lambda,$$
$$U^{-1}b_{\mathbf{q}\lambda}U = b_{\mathbf{q}\lambda} + \sqrt{n}\,\beta_\lambda \tag{10}$$

on the condensed-mode operators, one separates the transform of (1) as follows:

$$U^{-1}HU = H_0 + V$$
$$H_0 = W_0 + \sum_{\mathbf{k}} \left[\frac{\hbar^2k^2}{2m} a_{\mathbf{k}}{}^\dagger a_{\mathbf{k}} + \sum_\lambda \left(\hbar\omega_0 + \frac{\hbar^2k^2}{2m}\right) a_{\mathbf{k}\lambda}^\dagger a_{\mathbf{k}\lambda} + \sum_\lambda \hbar ck b_{\mathbf{k}\lambda}^\dagger b_{\mathbf{k}\lambda}\right] \tag{11}$$

where the thermal average of V in the ensemble determined by H_0 is negligible in the thermodynamic limit,

$$\frac{W_0}{n\hbar\omega_0} = \left(1 + \frac{\hbar\omega_0 z^2}{2mc^2}\right)\sum_\lambda |\alpha_\lambda|^2 + \left\{z + 2\pi\rho\alpha' z^{-1}\left[1 - \left(\frac{T}{T_c}\right)^{3/2}\right]\right\}\sum_\lambda |\beta_\lambda|^2$$
$$- (\pi\rho\alpha)^{1/2} z^{-1/2} \sum_\lambda (\alpha_\lambda{}^*\beta_\lambda\alpha_0 + \alpha_0{}^*\beta_\lambda{}^*\alpha_\lambda) \tag{12}$$

and $|\alpha_0|^2$ has been eliminated by the atom-number conservation condition

$$|\alpha_0|^2 + \sum_\lambda |\alpha_\lambda|^2 = 1 - \left(\frac{T}{T_c}\right)^{3/2} \tag{13}$$

Here T_c is the Bose–Einstein condensation temperature determined by the condition

$$\int \{[\exp(\beta_c\hbar^2k^2/2m) - 1]^{-1} + [\exp(\beta_c\hbar\omega_0)\exp(\beta_c\hbar^2k^2/2m) - 1]^{-1}\}\, d^3k = 8\pi^3\rho \tag{14}$$

with $\beta_c = (k_B T_c)^{-1}$; T_c differs from the usual ideal Bose gas condensation temperature only by an utterly negligible amount of order $T_c \exp(-\hbar\omega_0/k_B T_c)$, where $\hbar\omega_0/k_B T_c \sim 10^5$. The expressions (12) and (13) are obvious generalizations of (3) and (4). The free energy of H_0 is

$$\begin{aligned} F_0 = W_0 &+ (\Omega/8\pi^3\beta) \int \ln[1 - \exp(-\beta\hbar^2 k^2/2m)]\, d^3k \\ &+ (2\Omega/8\pi^3\beta) \int \ln[1 - \exp(-\beta\hbar\omega_0) \exp(-\beta\hbar^2 k^2/2m)]\, d^3k \\ &+ (2\Omega/8\pi^3\beta) \int \ln[1 - \exp(-\beta\hbar ck)]\, d^3k \end{aligned} \tag{15}$$

This is the Helmholtz free energy since the atom-number conservation condition has been introduced explicitly rather than indirectly via a chemical potential. The second term in (15) is the contribution of noncondensed ground-state atoms and plays an essential role in the temperature dependence. The third is the contribution of noncondensed excited-state atoms and is negligible at the temperatures of interest because of the aforementioned factor $\exp(-\hbar\omega_0/k_B T)$. The fourth term is the contribution of real photons and is likewise negligible. On the other hand, the coherent contributions of condensed virtually excited atoms and condensed virtual photons are important and are included in W_0.

The minimization of W_0 with respect to q (hence z), α_0, α_λ, and β_λ subject to the constraint (13) is carried out as was the minimization of (3), so we shall only state the results. If $\alpha < 2\alpha'$, the minimum occurs at $\alpha_\lambda = \beta_\lambda = 0$, corresponding to the normal condensed phase of the ideal Bose gas. If $\alpha > 2\alpha' > 0$, then the minimum occurs at nonzero values of the exciton condensate and photon condensate parameters, the minimum value of W_0 is

$$\frac{W_0}{n\hbar\omega_0} = -\frac{(\alpha - 2\alpha')^2}{8\alpha\alpha'} \left[1 - \left(\frac{T}{T_c}\right)^{3/2}\right] \tag{16}$$

and the condensate occupations are

$$\begin{aligned} \langle a_0{}^\dagger a_0 \rangle_0 &= n x_0{}^2 = n\, \frac{\alpha + 2\alpha'}{2\alpha} \left[1 - \left(\frac{T}{T_c}\right)^{3/2}\right] \\ \langle a_{\mathbf{q}1}^\dagger a_{\mathbf{q}1} \rangle_0 &= n x_1{}^2 = n\, \frac{\alpha - 2\alpha'}{2\alpha} \left[1 - \left(\frac{T}{T_c}\right)^{3/2}\right] \\ \langle b_{\mathbf{q}1}^\dagger b_{\mathbf{q}1} \rangle_0 &= z_{\min} n y_1{}^2 = n^{2/3}\, \frac{2\pi c \rho^{1/3}}{\omega_0}\, \frac{\alpha^2 - 4(\alpha')^2}{16\pi\rho\alpha(\alpha')^2} \end{aligned} \tag{17}$$

Note that the photon condensate occupation $\langle b_{\mathbf{q}1}^\dagger b_{\mathbf{q}1} \rangle_0$ for $0 < T < T_c$ is the same as the ground-state expression (9); accordingly, this occupation suffers a discontinuity at the transition temperature but then remains constant for

$0 \leqslant T < T_c$. On the other hand, the atomic ground-state condensate occupation $\langle a_0{}^\dagger a_0 \rangle_0$ and exciton condensate occupation $\langle a_{\mathbf{q}1}^\dagger a_{\mathbf{q}1} \rangle_0$ vary with temperature in the same way as the normal Bose condensate occupation of the ideal Bose gas. All of the occupations (17) are discontinuous at α, $\alpha' = 0$ if one "turns on" α and α' in such a way that their ratio remains constant.

If one drops the negligible second and third integrals in (15), then the Helmholtz free energy F_0 reduces to the sum of W_0 and the ideal Bose gas free energy. The various thermodynamic functions are then easily evaluated. For example, the specific heat at constant volume is found to be

$$\frac{C_V}{k_B} = -\frac{3}{32}\frac{\hbar\omega_0}{k_B T_c}\frac{(\alpha - 2\alpha')^2}{\alpha\alpha'}\left(\frac{T}{T_c}\right)^{1/2} + 1.925\left(\frac{T}{T_c}\right)^{3/2} \tag{18}$$

This is highly pathological, the negative term overwhelming the positive ideal Bose gas term since $\hbar\omega_0/k_B T_c \sim 10^5$. Similarly, the pressure is found to be negative and larger than the positive ideal Bose gas expression by a factor of order $\hbar\omega_0/k_B T_c$, and the ground-state energy and free energy themselves are much too negative ($\sim -\hbar\omega_0$). These pathological results signal the failure of our implicit assumption of a single homogeneous phase. In fact, the model (1) is unstable for $T < T_c$ against phase separation into a very high-density liquid phase and a low-density gas phase. It might be amusing to investigate this phase separation in detail, but we shall not do so, since such a collapse would not occur in a more realistic model including repulsive interactions. The actual binary collision matrix elements[6,7] are of the right order of magnitude to prevent such a collapse, but their consistent inclusion would require a more refined method than the simple variational treatment used here. For example, one might try to extend the Bogoliubov model by inclusion of appropriate atom–radiation interaction terms. The dipolar interaction should probably also be included explicitly (rather than as an effective potential between ground-state atoms), since it plays an important role in the theory of excitons, our crude treatment here already indicating the possible importance of excitonic effects. The repulsive interactions might appropriately be treated by a nonperturbative method, as, e.g., in the Bogoliubov model or the Lee–Huang–Yang theory of the hard-sphere Bose gas.

In view of the drastically simplified nature of the model (1), one should be cautious about drawing any firm conclusions with regard to real liquid ^{4}He, but we regard the results obtained here as a motivation for investigating the possibility of such superradiant effects in more realistic models. If one literally interprets α in (1) as the polarizability of the lowest P state (of excitation energy $\hbar\omega_0$), then the f-sum rule requires $\alpha < \alpha' = 2e^2/m_e\omega_0{}^2$, so that the condition $\alpha > 2\alpha'$ for superradiance is violated, in analogy with a previous observation[10] regarding the Dicke model. Such a conclusion does not, however, exclude the possibility of a superradiant transition in a more realistic model. The virtual transitions involved are highly nonresonant (involving photons of energy $\hbar c q_{\min} \ll \hbar\omega_0$), so that a realistic model should allow for excitation of all the higher excited states having nonzero dipole matrix elements to the ground state. These need not contribute to the Hamiltonian in the same way that they do to the f-sum rule, so that one cannot make a rigorous prediction of the relative contribution of the A and A^2 terms to a realistic model of superradiance on the basis of the f-sum rule.

If superradiance of a photon mode of macroscopic wavelength were to occur in real superfluid liquid ^{4}He (albeit in an attenuated form due to the presumably very small Bose-condensed fraction), it would provide an "effective field" to align the polarizations of virtually excited atoms, thus converting the atomic excited-state degeneracy into a discrete degeneracy of the macroscopic system of the type required for a correct treatment of the λ transition according to Tisza's theory.[1] Of course, it remains to be seen whether such a program can be successfully carried out.

Such superradiance in liquid ^{4}He might also lead to some bizarre effects not yet observed, e.g., an anomalous electromagnetic response in the neighborhood of the wavelength of the superradiant mode and a sensitivity of the λ anomaly in the specific heat to an externally applied field of the same wavelength (in the microwave region).

A superradiant transition involving nonlocalized atoms, of the type we have discussed, bears a considerable resemblance to the pion condensation transition, which has been investigated[11,12] as a possible phenomenon in neutron star matter and abnormally dense nuclear matter formed in high-energy collisions of heavy nuclei. It is amusing to note that also in that case the condensed phase was found to be unstable against collapse and this instability was attributed to an inadequate treatment of repulsive interactions. One essential difference is, however, that the pion condensation occurs at a finite q value rather than the infinitesimal value (corresponding to macroscopic wavelength) that we have found in our superradiance model. Furthermore, the presumed atomic Bose condensation in liquid ^{4}He furnishes a mechanism of enhancement of such a transition that is absent in a system of nucleons or other fermions. For this reason, superradiance is probably a more remote possibility in liquid ^{3}He, although it might conceivably occur since the Cooper pairs of ^{3}He atoms are believed to undergo condensations that have some similarity to Bose condensation.

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