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# Variational density-matrix theory of liquid $^4$He at nonzero temperatures

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We employ the minimum principle for the Helmholtz free energy to develop a self-consistent variational theory of liquid $^4$He at nonzero temperatures. Using the fact (demonstrated here) that the equilibrium density matrix of a boson system has non-negative matrix elements in coordinate representation, the trial density matrix in coordinate space is chosen to be an exponentiated sum of two-body functions. Adopting the separability assumption in conjunction with the hypernetted-chain approximation, we determine these functions optimally by a coupled set of Euler-Lagrange equations for the structure function $S(k,T)$ and the energy $\epsilon(k,T)$ of the elementary excitations. In a phenomenological study we analyze the variation of $S(k,T)$ and $\epsilon(k,T)$ with temperature, and discuss the relationship by employing experimental data on these functions at long wavelength and around the roton minimum.

The microscopic theory of the ground state of a strongly correlated boson fluid, i.e., liquid $^4$He at zero temperature, is well understood. The successful application of the "exact" Green's-function Monte Carlo method[1] to the simulation of the ground-state wave function by Whitlock, Ceperley, Chester, and Kalos[2] provides computer experimental information about the equation of state, liquid structure function $S(k)$, condensate fraction, and other useful quantities obtainable from the ground state. The parallel development of a functional representation of the ground state by a correlated wave function of the general form[3-6]

$$\Psi_0=\exp\left[\frac{1}{2}\left[\sum_{\substack{i,j=1\\ i<j}}^{N} u_2(r_{ij}) + \sum_{\substack{i,j,k=1\\ i<j<k}}^{N} u_3(\vec{r}_i,\vec{r}_j,\vec{r}_k)+\cdots\right]\right], \tag{1}$$

where the two- and three-body terms $u_2$ and $u_3$ are determined by Euler-Lagrange equations,[5,6]

$$\frac{\delta}{\delta u_2(r)}\frac{\langle\Psi_0|H|\Psi_0\rangle}{\langle\Psi_0|\Psi_0\rangle}=0\,,$$
$$\frac{\delta}{\delta u_3(\vec{r}_1,\vec{r}_2,\vec{r}_3)}\frac{\langle\Psi_0|H|\Psi_0\rangle}{\langle\Psi_0|\Psi_0\rangle}=0\,, \tag{2}$$

has provided a variational procedure capable of yielding quantitative agreement with the numerical results of the Green's-function Monte Carlo method of Ref. 2. Moreover, the analytic structure of the quantities $u_2$ and $u_3$ may be interpreted in terms of the zero-point motion of the elementary excitation spectrum of the fluid.[7]

Our present objective is to generalize the variational theory of liquid $^4$He to finite temperatures in such a way as to take advantage of the large body of work which has been acquired in the Jastrow-Euler-Lagrange method and its generalization.[8] While most of the theoretical understanding of the temperature dependence of $^4$He has been based on quasiparticle models of the Hamiltonian $H$, we will preserve the exact form of $H$ and instead concentrate on approximations for the density matrix in the coordinate-space representation. Thermodynamic functions of interest can then be calculated from the Helmholtz free energy, while the density matrix will be used to calculate such quantities as the liquid structure function and condensate fraction at nonzero temperatures.

At finite temperature, $T=(k_B\beta)^{-1}$, variational theory appropriately begins with the Gibbs-Delbrück-Moliere minimum principle[9] for the Helmholtz free energy $F$,

$$F\le \mathrm{Tr}\left[W_tH+\frac{1}{\beta}W_t\ln W_t\right]\equiv F_t\,, \tag{3}$$

where $W_t$ is a trial statistical operator of suitable form on the Hilbert space of the Hamiltonian $H$ with appropriate (Bose) statistics. The operator $W_t$ must be normalized, $\mathrm{Tr}\,W_t=1$, and must be positive definite on the Hilbert space. The equality sign in Eq. (3) is obtained only if $W_t$ is the equilibrium density matrix $W=e^{-\beta H}/\mathrm{Tr}\,e^{-\beta H}$. In order that the thermodynamic and statistical mechanical definitions of derived thermodynamic functions such as entropy and internal energy be self-consistent, the trial density matrix must be properly restructured to have no explicit dependence upon temperature.[10]

To avail ourselves of the results and methods developed in the ground-state variational calculations of the helium liquids, we express the right-hand side of expression (3) in the coordinate-space representation. In that case, $W_t(\vec r_1,\ldots,\vec r_N;\vec r\,'_1,\ldots,\vec r\,'_N)$ is the trial $N$-body density matrix for a system of $N$ particles. There are two useful properties of the exact density matrix which we require for the trial functions: positive coordinate-space representation and product-cluster property. The latter is obtained from the same property of many-body wave functions for fluids, and implies, in particular, that the density matrix can be written as

$$W_t(\vec r_1,\ldots,\vec r_N;\vec r\,'_1,\ldots,\vec r\,'_N) =\Phi^*(\vec r_1,\ldots,\vec r_N)Q(\vec r_1,\ldots,\vec r_N;\vec r\,'_1,\ldots,\vec r\,'_N) \times\Phi(\vec r\,'_1,\ldots,\vec r\,'_N)\ , \tag{4}$$

where the incoherence factor $Q$ contains no factor depending only on primed or unprimed coordinates alone. Of course, if $W_t$ is the density matrix of a single state, then $Q=1$, and $\Phi$ is the wave function of that state. The fact that the operator $W_t$ in the coordinate-space representation has non-negative matrix elements depends only on the Bose statistics of the system (see the Appendixes for details). As a consequence of these two properties, the density matrix may be represented as an exponentiated sum of two-body, three-body, . . . , multibody real functions in analogy to the Feenberg form of the boson ground-state wave function (1).[5]

In a first step within our approach based on the variational principle (3), we adopt a trial density matrix with the components

$$\Phi=\Psi_0(\vec r_1,\ldots,\vec r_N)\prod_{\substack{i,j=1\\ i<j}}^{N}\exp\tfrac{1}{2}\Delta u(r_{ij})\ , \tag{5}$$

$$Q=\prod_{i,j=1}^{N}\exp\gamma(\,|\,\vec r_i-\vec r\,'_j\,|\,)\ , \tag{6}$$

which has the required structure discussed above. The wave function $\Psi_0$ represents the ground state of liquid $^4$He (or some approximation to it), the two-body function $\Delta u(r)$ describes the finite-temperature shift of the "pseudopotential" $u_2(r)$, Eq. (1), and the quantity $\gamma(r)$ generates the incoherent part of the density matrix. The explicit forms (5) and (6) have been originally derived as an approximation to the density matrix at temperatures far below the $\lambda$ point by approximately incorporating the effects of noninteracting phonons,[11] or Bijl-Feynman excitations.[12] Expressions (4)–(6) for the density matrix also have been used in recent studies of the temperature dependence of spatial correlations in $^4$He by Reatto and coworkers.[13,14]

The *Ansätze* (5) and (6) represent the simplest class of trial density matrices appropriate for liquid $^4$He, particularly when the wave function $\Psi_0$ is approximated by the Jastrow form described by Eq. (1) with $u_n=0$ for $n>2$, so that

$$W_t(\vec r_1,\ldots,\vec r_N;\vec r\,'_1,\ldots,\vec r\,'_N) =Z_t^{-1}\prod_{\substack{i,j=1\\ i<j}}^{N}\exp[\tfrac{1}{2}u(r_{ij})] \times\prod_{i,j=1}^{N}\exp\gamma(\,|\,\vec r_i-\vec r\,'_j\,|\,) \times\prod_{\substack{i,j=1\\ i<j}}^{N}\exp[\tfrac{1}{2}u(r'_{ij})]\ , \tag{7}$$

with $Z_t$ being the associated trial partition function. It should be noted that *Ansatz* (7) provides an admissible set of trial density matrices possessing non-negative eigenvalues provided that the function $\gamma(r)$ has a semipositive Fourier inverse (see the Appendixes for details). As a next step we optimize the free-energy functional $F_t$, expression (3), at a given nonzero temperature, thus determining the functions $u(r)$ and $\gamma(r)$ of Ansatz (7) from the corresponding Euler-Lagrange equations:

$$\frac{\delta F_t}{\delta\gamma(r)}=0\ , \tag{8}$$

$$\frac{\delta F_t}{\delta u(r)}=0\ . \tag{9}$$

We emphasize that the optimization procedure ensures a self-consistent evaluation of thermodynamic quantities, i.e., thermodynamic and statistical-mechanical definitions of observables such as the entropy lead to the same formal expressions. In this important aspect our approach improves upon the earlier work of Refs. 11–14. The Euler-Lagrange equations (8) and (9) constitute a coupled set of equations for functions $u(r)$ and $\gamma(r)$. Since the entropy term contributing to the trial free energy $F_t$ varies with temperature, the optimal solutions of Eqs. (8) and (9) depend on the temperature. It is straightforward to show that in the limit $T\rightarrow 0$ the phonon density matrix used in Refs. 11–14 is a solution of the Euler-Lagrange equations (8) and (9) with $\Psi_0$ corresponding to the optimum Jastrow ground-state trial function. Deviations from the low-temperature results must occur as quasiparticle correlations become more important. These are accounted for in part by the optimization procedure at a fixed nonzero temperature based on the trial matrix (7), although it might be necessary to use a more elaborate choice for $W_t$ in order to obtain quantitative agreement with the experimental results on the elementary excitation energies as the temperature increases.

Employing Ansatz (7) as input, the first term in expression (3), which represents the internal energy $U_t$, can be evaluated in a standard manner. Using the Jackson-Feenberg form for the kinetic energy, we obtain

$$U_t=N\frac{\rho}{2}\int v^*(r)g(r)d\vec r+\frac{N}{(2\pi)^3\rho}\int\frac{\hbar^2k^2}{2m}\tilde\gamma(k)d\vec k\ , \tag{10}$$

where

$$v^*(r)=v(r)-\frac{\hbar^2}{4m}\vec\nabla^2[u(r)+2\gamma(r)]\ . \tag{11}$$

Equation (10) involves the radial distribution function $g(r)$ associated with the "pseudopotential" $u(r)+2\gamma(r)$, $\tilde{\gamma}(k)$ is the dimensionless Fourier inverse of the incoherence function $\gamma(r)$, and $\rho$ is the number density. The potential $v(r)$ in expression (11) describes the bare interaction between two helium atoms. Standard hypernetted-chain equations provide the relationship between the radial distribution function $g(r)$ and the quantity $u(r)+2\gamma(r)$.

The entropy of the trial density matrix, defined as

$$S_t=-k_B\mathrm{Tr}(W_t\ln W_t)\ , \tag{12}$$

provides a difficult challenge to calculate as a functional of quantities $u$ and $\gamma$. However, we can evaluate $S_t$ if we adopt a generalized separability approximation[15] or, equivalently, a cumulant analysis in the density-fluctuation operators.[16] The introduction of an approximation, of course, invalidates the variational theorem in principle. However, our experience with this approximation in other calculations gives us confidence that it produces an excellent approximation to the exact Euler-Lagrange equation. Furthermore, the result is exact if $\gamma(r)$ contains only one Fourier coefficient and its inverse ($\vec{k}$ and $-\vec{k}$). Thus the approximation amounts to ignoring interference effects between different momentum-space components. The analysis of $S_t$ has been performed in detail in Ref. 17 and obtains an approximation for the entropy (12) as a single sum over wave numbers. The explicit form of the result is consistent with a change of variables to produce an elementary excitation structure for the entropy,

$$S_t=\frac{N}{(2\pi)^3\rho}\int\{[1+n(k)]\ln[1+n(k)]-n(k)\ln n(k)\}d\vec{k}\ . \tag{13}$$

In this approximation the incoherence term $\tilde{\gamma}(k)$ of *Ansatz* (7) must be considered to be a functional of quantities $n(k)$ and $S(k)$ through the relationship[17]

$$\tilde{\gamma}(k)=\frac{1}{S(k)}n(k)[1+n(k)]\ , \tag{14}$$

where $S(k)$, the liquid structure function at nonzero temperatures, is 1 plus the dimensionless Fourier inverse of $g(r)-1$. The same functional relationship has been employed in earlier references, for example, in Refs. 11 and 12, where function $n(k)$ appears as the average boson occupation probability for temperature-independent quasiparticle excitations. Empirical justification for the form (13) of the entropy is obtained from the thermodynamics analysis of Bendt, Cowan and, Yarnell,[18] who find that quasiparticle statistical mechanics, together with the measured elementary excitation spectrum, provide a good description of $^4$He to $T=1.95$ K. The usefulness of the separability assumption, inherent in Eqs. (13) and (14), at high temperatures, remains to be explored.

The variational calculation of expressions (8) and (9) at a fixed temperature based on Eqs. (10), (11), (13), and (14) is most conveniently done after the change of variables from the pair $\gamma(r)$ and $u(r)$ to the pair $n(k)$ and $g(r)$ or $S(k)$. Variation of the trial free energy $F_t=U_t-TS_t$, with respect to quantity $n(k)$ holding the distribution function $g(r)$ fixed, gives the optimization condition

$$\epsilon(k)=\frac{\hbar^2k^2}{2mS(k)}\coth\left[\frac{\beta}{2}\epsilon(k)\right]\ , \tag{15}$$

where we have introduced a function $\epsilon(k)$ defined by

$$n(k)=(e^{\beta\epsilon(k)}-1)^{-1}\ . \tag{16}$$

Variation of the functional $F_t$ with respect to the radial distribution function at fixed quantity $n(k)$ yields a Euler-Lagrange equation of the form

$$\left[-\frac{\hbar^2}{m}\nabla^2+\hat{v}(r)\right]\sqrt{g(r)}=0\ . \tag{17}$$

Here, the effective potential $\hat{v}(r)$ is defined by

$$\hat{v}(r)=v(r)+w(r)+\phi(r)\ , \tag{18}$$

with the components

$$w(r)=\frac{-1}{(2\pi)^3\rho}\int\frac{\hbar^2k^2}{4m}\frac{[2S(k)+1][S(k)-1]^2}{S^2(k)}e^{i\vec{k}\cdot\vec{r}}d^3k\ , \tag{19}$$

$$\phi(r)=\frac{-1}{(2\pi)^3\rho}\int\frac{\hbar^2k^2}{m}\frac{n(k)[1+n(k)]}{S^2(k)}e^{i\vec{k}\cdot\vec{r}}d^3k\ . \tag{20}$$

At zero temperature the function $n(k)$ vanishes. Thus, we have $\phi(r)=0$ and the Euler-Lagrange equation (17) reduces to that found by Lantto and Siemens for the optimal structure function associated with a ground-state wave function of Jastrow type.[19]

The solutions of the two coupled Euler-Lagrange equations (16) and (17) depend upon the temperature $T$, which will hereafter be included as an explicit argument for clarity. In particular, the relation (15) between the solutions $\epsilon(k,T)$ and $S(k,T)$ reads

$$\epsilon(k,T)\tanh\left[\frac{\epsilon(k,T)}{2k_BT}\right]=\frac{\hbar^2k^2}{2m}\frac{1}{S(k,T)}\ . \tag{21}$$

It should be noted that this result is equivalent to that obtained from a single resonance *Ansatz* for the density-density response function, together with the $f$-sum rule and the fluctuation-dissipation theorem, to produce a relationship between the excitation energy in the *Ansatz* and the liquid structure function. This feature is well known from the $T=0$ Jastrow Euler-Lagrange theory; the fact that it generalizes to finite temperatures with our model density matrix serves to emphasize the physical content of the model, namely the absence of backflow contributions to the density matrix. As in the $T=0$ case, the equivalence of these two approaches requires that one use the Euler-Lagrange solution; a parametrized density matrix would not produce the simple result of Eq. (21).

In the limit that $T\rightarrow 0$ we recover from Eq. (21) the familiar Bijl-Feynman result,

$$\epsilon(k,0)=\frac{\hbar^2k^2}{2m}\frac{1}{S(k,0)}\ . \tag{22}$$

In this case the function $u(r)$ is long ranged, $u(r)\sim r^{-2}$

as $r \rightarrow \infty$, and the associated structure function vanishes linearly with wave vector $k$, so that the elementary excitation energy $\epsilon(k,0)$ has the correct linear $k$ behavior. At finite temperatures the effective pseudopotential $u(r,T)+2\gamma(r,T)$ is short ranged, falling off at least as $r^{-4}$, thus generating a finite value of $S(0,T)$ proportional to the temperature,

$$S(0,T)=k_B T/mc^2 \; . \tag{23}$$

Equation (23) defines a temperature-dependent quantity $c$ which is related to the isothermal compressibility $K_T$ by $mc^2=K_T^{-1}$. This notation is motivated by noting that the long-wavelength limit of relation (21) together with Eq. (23) indicates that function $\epsilon(k,T)$ is linear in the momentum $\hbar k$ with slope $c$,

$$\epsilon(k,T) \rightarrow \hbar ck \quad \text{as } k \rightarrow 0 \; . \tag{24}$$

Result (24) and expression (16) with the optimal function $\epsilon$, i.e.,

$$n(k,T)=\frac{1}{e^{\epsilon(k,T)/k_B T}-1} \; , \tag{25}$$

suggests that we may interpret the solution $\epsilon(k,T)$ as the temperature-dependent generalization of the Bijl-Feynman excitation spectrum.[20] Consequently, the factor $c$ in Eq. (24) may be viewed as the temperature-dependent velocity of the long-wavelength excitations.

In the following we will concentrate on a phenomenological study of the relationship (21) between the excitation energies $\epsilon(k,T)$ and the liquid structure function $S(k,T)$. To study the variation with temperature, we separately consider the behavior of the density fluctuations at small momentum transfer and that of rotons. Our phenomenological analysis of Eq. (21) at small wave numbers is based on the experimentally measured structure functions $S(k,T)$ of Hallock[21] and Svensson, Sears, Woods, and Martel.[22,23] Employing those experimental results for temperatures $T<T_\lambda=2.17$ K and wave numbers $k \lesssim 0.4$ Å$^{-1}$, Eq. (21) yields associated excitation energies $\epsilon(k,T)$, which indeed agree very well with the experimentally determined excitation energies of Cowley and Woods[24] at low momentum (Fig. 1). In contrast, the theoretically inconsistent application of the Bijl-Feynman relation (22) at nonzero temperature leads to an approximation for the excitation energies which violates the linear dependence at small momenta and disagrees with experimental results (curves 1—3 in Fig. 1). The differences between the predictions of Eqs. (21) and (22) increase markedly with increasing temperature, as can be noted by comparing the 3-K results in Fig. 1. Note also that the temperature-dependent effects at 1 K are missing beyond $k \gtrsim 0.3$ Å$^{-1}$, where the results of Eqs. (21) and (22) merge.[25]

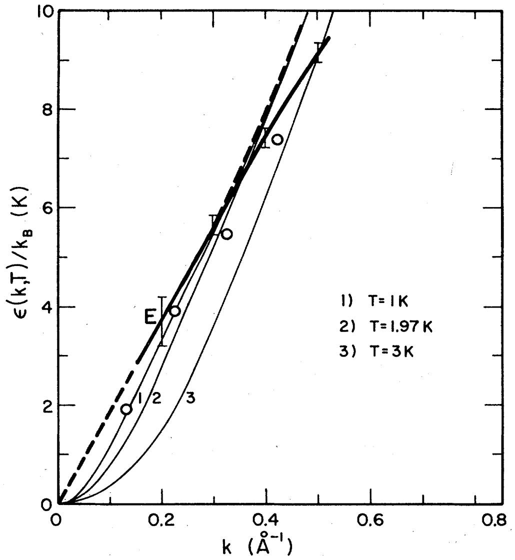

FIG. 1. Energy $\epsilon(k,T)$ of elementary excitations at small wave numbers $k$. Curve $E$ with the error bars represents experimental data of Ref. 24. Our theoretical results based on Eq. (21) and experimental structure functions for $T<T_\lambda$ are shown as a dashed line at $T=1$ K, and by ○ at $T=3.3$ K. Curves 1—3 depict results on the inconsistent application of the Bijl-Feynman excitation spectrum using finite-temperature structure factors, thus violating the linear momentum dependence of $\epsilon(k,T)$.

In Fig. 2 we display in detail the temperature dependence of the excitation energies calculated from Eq. (21) using the "smoothed" experimental data of Ref. 23 for $S(k,T)$ at saturated vapor pressure and $k \lesssim 0.5$ Å$^{-1}$. The data are exhibited as the phase velocity (multiplied by $\hbar$). For reference we have included the experimental isothermal sound velocity,[28] which should be related to the remaining data by

$$\lim_{k\rightarrow 0}[\epsilon(k)/k]=\hbar c \; .$$

This relationship is clearly satisfied even at temperatures well above $T_\lambda$. However, that will always be the case as

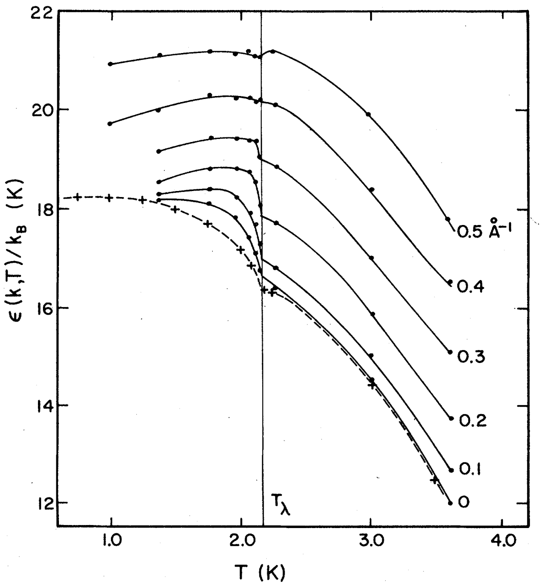

FIG. 2. Phase velocity $\epsilon(k,T)/k$ of elementary excitations at several wave numbers as a function of temperature, using Eq. (21) with input structure functions of Ref. 23. The curve labeled zero is $\hbar c$, where $c$ is the isothermal sound velocity as determined by the theory. The dashed curve is the experimental result from Ref. 28.

long as Eq. (23) is satisfied, as it must be in the present phenomenological application of the theory since $S(k,T)$ is actually taken from experiment. Similarly, in the case of the low-temperature, lowest-wave-number results, the fact that this limit seems to converge to a somewhat different constant than the thermodynamic value of $\hbar c$ is evidently a reflection of the fact that the combination of systematic and statistical errors in the small-$k$ measurements of $S(k)$ is greater than the other values displayed, and is sufficiently large to encompass the proper isothermal sound-velocity result.[21]

The calculated phase velocity shown in Fig. 2 increases slightly with increasing temperature up to about 1.9 K and then decreases rather rapidly until the $\lambda$ transition. Some additional experimental support for this behavior is given by the measurements of Cowley and Woods[24] on the mean energy of the scattered total neutron groups (see Sec. III C and particularly Fig. 16 of Ref. 29).

Recent elaborate measurements on the structure function $S(k,T)$ in the region of roton momenta have revealed that the principal peak increases and sharpens as the temperature approaches the $\lambda$ temperature from below.[29,30] At such wave numbers, Eq. (21) does not give an appropriate description of the relationship between the structure function and the excitation energies since backflow effects and quasiparticle-interaction effects are completely ignored in *Ansatz* (7). At wave numbers $k>0.6$ Å$^{-1}$ and temperatures $T<T_\lambda$, Eq. (21) reduces to the familiar Bijl-Feynman relation, yielding theoretical roton energies which are 2—3 times larger than the observed energies. Nevertheless, the present approximation may give some qualitative insight on the sharpening of the liquid-structure-factor peak. We rewrite Eq. (21) as an equation for the structure function in terms of the temperature-dependent excitation spectrum,

$$S(k,T)=\frac{\epsilon(k,0)S(k,0)}{\epsilon(k,T)}\coth\left[\frac{\epsilon(k,T)}{2k_BT}\right]. \qquad (26)$$

Earlier expressions for $S(k,T)$ set, effectively, $\epsilon(k,T)\approx\epsilon(k,0)$, thus neglecting the temperature dependence of the spectrum everywhere in Eq. (26),

$$S(k,T)\approx S(k,0)\coth\left[\frac{\epsilon(k,0)}{2k_BT}\right]. \qquad (27)$$

This approximation results merely in an increase in the structure function at every value of momentum and thus no sharpening. In contrast, Eq. (26) tells us that we may trace some of the observed effect to an appropriate sharpening of the relative shift in the excitation energies by varying the temperature.

Neutron-scattering experiments which give information on the energy shift around the roton minimum have been carried out at several temperatures.[24,31-33] To learn whether the experimental information gives some qualitative support to the above discussion we cast Eq. (26) into the slightly more general form

$$\frac{S(k,T_2)}{S(k,T_1)}=\frac{\epsilon(k,T_1)}{\epsilon(k,T_2)}\frac{\tanh[\epsilon(k,T_1)/2k_BT_1]}{\tanh[\epsilon(k,T_2)/2k_BT_2]}, \qquad (28)$$

and employ the smoothed data of Ref. 23 on the structure function at temperatures $T_1=1$ K and $T_2=2.15$ K together with the experimental excitation energies[24] at $T_1=1$ K to evaluate the form factor $\epsilon(k,T_1)/\epsilon(k,T_2)$.

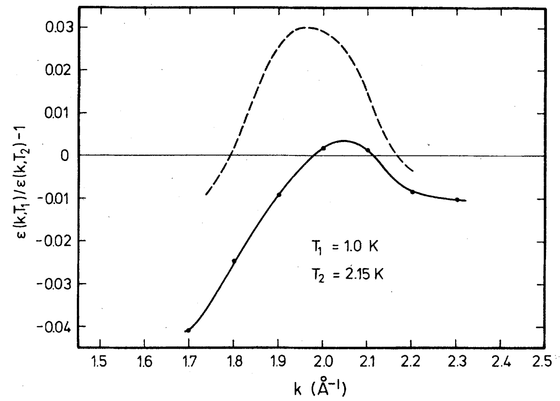

FIG. 3. Form factor $\epsilon(k,T_1)/\epsilon(k,T_2)-1$ in the region of roton wave numbers. Solid curve gives results of Eq. (28) with input data on $S(k,T)$ at $T_1=1$ K and $T_2=2.15$ K taken from Ref. 23 and on $S(k,T)$ at $T=1$ K of Ref. 24. Experimental data of Ref. 25 yield a more pronounced profile, qualitatively indicated by the dashed line.

The results of this analysis are shown in Fig. 3 by the solid line. We find, indeed, a positive maximum at $k\sim2.05$ Å$^{-1}$ and a negative profile at $k<1.95$ Å$^{-1}$ and $k>2.1$ Å$^{-1}$ for the relative shift $\epsilon(k,T_1)/\epsilon(k,T_2)-1$. However, the peak is extremely small leading to an unrealistically small roton-energy shift, $\epsilon(k,T_1)-\epsilon(k,T_2)$, of only 0.03 K. Using, instead, the experimental information of Ref. 25 on the temperature dependence of the maximum height of the principal structure-function peak

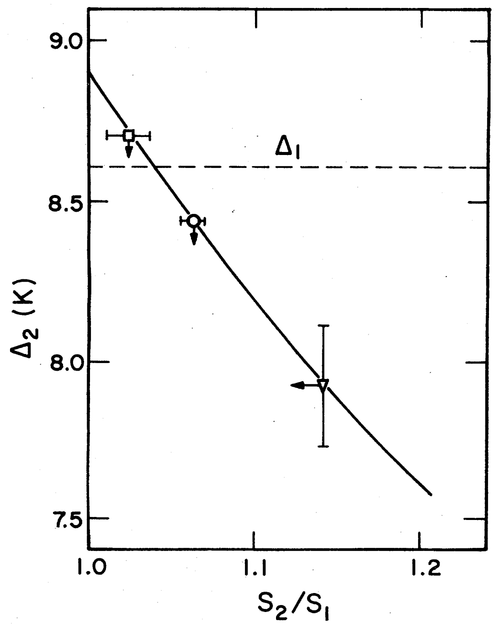

FIG. 4. Temperature dependence of the roton gap $\Delta$ as a function of the structure-factor ratio as obtained from Eq. (28). □ gives $S_2/S_1$ from neutron scattering, Ref. 23; ○ gives $S_2/S_1$ from x-ray scattering, Ref. 29; △ gives the measured gap at $T=2.15$ K, Ref. 31.

at density $\rho = 150.3$ kg m$^{-3}$, the peak of the energy form factor (minus 1) increases roughly about 1 order of magnitude (dashed line, Fig. 3). Consequently, the theoretical roton-energy shift is about 0.3 K, which we may compare with an experimental energy shift of about 1 K reported by Mezei.[32] This order-of-magnitude difference in the predicted shift comes about because the percentage increase in $S(k)$ as one goes from 1 to 2.15 K is 3% in the neutron-diffraction measurements,[22] but is 6% in the x-ray measurements.[29] Since Eq. (28) depends only on the ratio of $S$ at the two temperatures, in Fig. 4 we show the roton gap $\Delta(T)$ as a function of this ratio as calculated in Eq. (28). $\Delta(T_1)$ is taken from Mezei's results at $T_1 = 1.0$ K. Shown on the curve are the experimental structure-function ratios at 1.9 Å$^{-1}$,[31] and the experimental value for $\Delta$ at $T_2 = 2.15$ K. Thus we see that, if Eq. (28) were to account for the entire shift in the roton minimum, the corresponding increase in the structure factor would have to be 14%, more than twice the largest observed shift. We do not suggest that such a large increase in $S(k)$ should have been observed. Instead, our results show that the thermal occupation of rotons and the induced correlations as manifested in the incoherence function $\gamma(r)$ can only account for a fraction of the shift in the roton minimum. The majority of the shift is certainly due to roton-roton interactions, which is the usual interpretation.[31]

In conclusion, the approach we have presented here should be considered as the initial step in a systematic development of a variational microscopic theory of a many-boson system at nonzero temperatures. To achieve a satisfactory description of liquid $^4$He at $T > 0$ we must, at least, (i) incorporate the effects of backflow and quasiparticle interactions by generalizing the *Ansatz* (7), and (ii) find better approximations for the entropy by improving upon the separability assumption.

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## APPENDIX A

Here we wish to demonstrate one result used in the above analysis: that the density matrix for bosons in thermal equilibrium has non-negative matrix elements in the coordinate-space representation.

The positivity of the coordinate-space matrix elements of the equilibrium boson density matrix follows by inspection from the path-integral representation of that quantity. It is well known that the Boltzmann density matrix has this property since it is the path integral of an exponentiated real function:

$$W_{\text{Boltzmann}}(R,R';\beta) = \int_{r_1}^{r_1'} \cdots \int_{r_N}^{r_N'} \exp - \left[ \frac{m}{2\hbar} \sum_i \int_0^{\beta\hbar} \dot{\vec{r}}_i''(t)^2 dt + \frac{1}{2\hbar} \sum_{\substack{i,j=1 \\ i \neq j}} \int_0^{\beta\hbar} V[\vec{r}_i''(t) - \vec{r}_j''(t)] dt \right] \mathscr{D}^N \vec{r}''(t) .$$

The corresponding expression for some other statistics is obtained by projection on this operator with an appropriate permutation sum. In particular, for boson statistics this is just the sum over permutations of the primed (*or* unprimed) coordinates:

$$W_{\text{Bose}}(R,R';\beta) = \frac{1}{N!} \sum_P W_{\text{Boltzmann}}(R,PR';\beta)$$

(see, for example, Eq. 10.77 of Ref. 34). Since this is the sum of terms each of which is non-negative, it too is non-negative.

## APPENDIX B

Here we will now demonstrate the second result: that *Ansatz* (7) represents a trial matrix which possesses the required property of having non-negative eigenvalues if the Fourier inverse of the function $\gamma(r)$ is non-negative.

The trial *Ansatz* (7) for the equilibrium density matrix of identical bosons in the coordinate-space representation is of the form

$$W_t(R,R') = \Phi^*(R) \prod_{i,j} e^{\gamma(|\vec{r}_i - \vec{r}_j'|)} \Phi(R') . \qquad \text{(B1)}$$

To prove that this provides a set of trial density matrices which satisfy the requirement that the eigenvalues be non-negative, we demonstrate that

$$\langle \psi | W_t | \psi \rangle \geq 0 \qquad \text{(B2)}$$

for any state $\psi$ if the Fourier inverse of function $\gamma(r)$ is non-negative. The proof follows immediately by introducing the Fourier transform into the incoherence factor:

$$\prod_{i,j} e^{\gamma(|\vec{r}_i - \vec{r}_j'|)} = \exp\left[ \frac{1}{N} \sum_k \tilde{\gamma}(k) \rho_k(R) \rho_k^*(R') \right] , \qquad \text{(B3)}$$

where the density-fluctuation operator $\rho_k = \sum_i e^{i\vec{k}\cdot\vec{r}_i}$. Inserting this expression into (B2), expanding the exponential in the incoherence factor, and collecting the primed and unprimed coordinates into separate factors, one arrives at the series

$$\begin{aligned}\langle\psi\,|\,W_t\,|\,\psi\rangle&=\int d^{3N}R\,d^{3N}R'\psi^*(R)W_t(R,R')\psi(R')\\&=\sum_{n=0}^{\infty}\frac{1}{n!}\frac{1}{N^n}\sum_{k_1,\ldots,k_n}\tilde{\gamma}(k_1)\cdots\tilde{\gamma}(k_n)\left|\int d^{3N}R\,\psi^*(R)\Phi^*(R)\rho_{k_1}(R)\cdots\rho_{k_n}(R)\right|^2 .\end{aligned}\tag{B4}$$

Thus for any state $\psi$ we find that (B2) is satisfied if $\tilde{\gamma}(k)\geq 0$.

Although this condition is sufficient, it has not been shown that it is necessary. To demonstrate the latter in our present calculation, we make a specific choice of $\psi$ in (B2), namely

$$\psi(R)=\rho_k(R) . \tag{B5}$$

Since the density operator is a symmetric function, this choice is applicable only to boson statistics. The expectation value we wish to examine is generated by the integral of $W_t$ over primed and unprimed coordinates:

$$\langle\rho_k\,|\,W_t\,|\,\rho_k\rangle=\frac{1}{2}\frac{d}{d\tilde{\gamma}_k}\int d^{3N}R\,d^{3N}R'W_t(R,R') . \tag{B6}$$

If one employs a Jastrow choice (or any of its generalizations) for $\Phi$, the integral can be evaluated using a cumulant analysis.[16] To apply this to the results in the text we adopt the same truncation of the cumulant analysis which was used above, namely the generalized separability approximation.[15] In that case we make use of (B6) to find that

$$\langle\rho_k\,|\,W_t\,|\,\rho_k\rangle=\frac{\tilde{\gamma}_k\,|S(k)|^2}{[1-\tilde{\gamma}_k^2\,|S(k)|^2]^2} , \tag{B7}$$

where $S(k)$ is a positive function for a positive semidefinite choice of $\Phi$ as we require here, defined by

$$S(k)=\frac{1}{N}\int\Phi(R)\,|\rho_k|^2d^{3N}R .$$

Clearly, the right-hand side of (B7) has the same sign as $\tilde{\gamma}(k)$, which completes the demonstration that $\tilde{\gamma}(k)$ must be non-negative. Although this part of the proof is weakened by the necessity to invoke an approximation, the important feature is that it is internally consistent with the approximation used to obtain Eq. (13). While it may turn out that interference effects produce a correction to (B6) which makes it positive for negative $\tilde{\gamma}(k)$, this would not constitute a proof that such negative values are permissible. That would require an examination of all possible choices of $\psi$. It is implausible that one cannot construct a choice for $\psi$ that would make (B2) negative when $\tilde{\gamma}(k)$ is negative.

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[1] M. H. Kalos, Nucl. Phys. **A328**, 153 (1979).

[2] P. A. Whitlock, D. M. Ceperley, G. V. Chester, and M. H. Kalos, Phys. Rev. B **19**, 5598 (1979).

[3] C. W. Woo, Phys. Rev. Lett. **28**, 1442 (1972).

[4] C. W. Woo and R. L. Coldwell, Phys. Rev. Lett. **29**, 1062 (1972).

[5] C. E. Campbell, Phys. Lett. **44A**, 471 (1973).

[6] C. C. Chang and C. E. Campbell, Phys. Rev. B **15**, 4238 (1977).

[7] C. De Michelis, G. Masserini, and L. Reatto, Phys. Rev. A **18**, 296 (1978).

[8] C. E. Campbell and F. J. Pinski, Nucl. Phys. **A328**, 210 (1979).

[9] A. Huber, in *Methods and Problems of Theoretical Physics*, edited by J. E. Bowcock (North-Holland, Amsterdam, 1970), Vol. 37.

[10] F. Wagner and H. Koppe, Z. Naturforsch. Teil A **20**, 1553 (1965).

[11] L. Reatto and G. V. Chester, Phys. Rev. **155**, 88 (1967).

[12] E. Feenberg, Ann. Phys. (N.Y.) **70**, 133 (1972).

[13] C. De Michelis, G. L. Masserini, and L. Reatto, Phys. Lett. **66A**, 484 (1978).

[14] G. Gaglione, G. L. Masserini, and L. Reatto, Phys. Rev. B **23**, 1129 (1981).

[15] C. E. Campbell and E. Feenberg, Phys. Rev. **188**, 396 (1969).

[16] C. E. Campbell, J. Math. Phys. **16**, 1076 (1975).

[17] G. Senger, Diplomarbeit 1983; and unpublished.

[18] P. J. Bendt, R. D. Cowan, and J. L. Yarnell, Phys. Rev. **113**, 1386 (1959).

[19] P. J. Siemens and L. J. Lantto, Phys. Lett. **68B**, 308 (1977).

[20] D. Pines, in *Many-Body Theory*, edited by R. Kubo (Benjamin, New York, 1966), Vol. 36.

[21] R. B. Hallock, Phys. Rev. A **5**, 320 (1972); and private communication.

[22] E. C. Svensson, V. F. Sears, A. D. B. Woods, and P. Martel, Phys. Rev. B **21**, 3638 (1980).

[23] V. F. Sears, E. C. Svensson, A. D. B. Woods, and P. Martel, Atomic Energy of Canada Ltd., AECL Report No. 6779 (1979) (unpublished).

[24] R. A. Cowley and A. D. B. Woods, Can. J. Phys. **49**, 177 (1971).

[25] This happens to be the momentum at which backflow corrections to the Bijl-Feynman theory and our temperature-dependent generalization become important. For $k\geq 0.3$ Å$^{-1}$ these anharmonic effects improve agreement with experiment in previous $T=0$ calculations (Refs. 26 and 27) and should do likewise at finite temperatures.

[26] D. K. Lee and F. J. Lee, Phys. Rev. B **11**, 4318 (1975).

[27] C. C. Chang and C. E. Campbell, Phys. Rev. B **13**, 3779 (1976); C. E. Campbell and F. J. Pinski, Nucl. Phys. **A328**, 210 (1979).

[28] J. S. Brooks and R. J. Donnelly, J. Phys. Chem. Ref. Data **6**, 51 (1977); R. D. McCarty, *ibid.* **2**, 923 (1973).

[29] H. N. Robkoff, D. A. Ewen, and R. B. Hallock, Phys. Rev. Lett. **43**, 2006 (79); F. W. Wirth, D. A. Ewen, and R. B. Hallock, Phys. Rev. B **27**, 5530 (1983).

[30] E. C. Svensson and A. F. Murray, Physica (Utrecht) **108B**, 1317 (1981).

[31] D. G. Henshaw and A. D. B. Woods, Phys. Rev. **121**, 1266 (1961).

[32] F. Mezei, Phys. Rev. Lett. **44**, 1601 (1980).

[33] A. D. B. Woods and E. C. Svensson, Phys. Rev. Lett. **41**, 974 (1978).

[34] R. P. Feynman and A. R. Hibbs, *Quantum Mechanics and Path Integrals* (McGraw-Hill, New York, 1965).