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Theory of the temperature dependence of spatial correlations in liquid ^{4}He

G. Gaglione and G. L. Masserini
Instituto di Fisica dell'Università and Gruppo Nazionale di Struttura della Materia del Consiglio Nazionale delle Ricerche, via Celoria 16, Milano, Italy

L. Reatto
Instituto di Fisica dell'Università di Parma and Gruppo Nazionale di Struttura della Materia del Consiglio Nazionale delle Ricerche, Instituto di Fisica, Università di Milano, Italy

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The temperature dependence of the structure factor $S(k,T)$ of liquid ^{4}He is studied on the basis of a model density matrix based on the elementary excitation picture and with roton excitations described in terms of the Feynman wave function. $S(k,T)$ is constructed starting from the experimental low-temperature $S(k)$, the energy spectrum $\epsilon_k(T)$, and the linewidth $\delta_k(T)$. No assumption is needed on the form of the Hamiltonian. The main peak of $S(k,T)$ has an anomalous behavior, it increases its height and becomes sharper as the temperature is increased; this is due to thermal population of roton states that causes an enhancement of the short-range order. This anomalous behavior ends slightly above T_λ when rotons become overdamped. Our result is in good quantitative agreement with experiment. This temperature effect becomes stronger at high density.

I. INTRODUCTION

The temperature dependence of the radial distribution function $g(r,T)$ and of the related structure factor $S(k,T)$ of liquid ^{4}He has recently attracted much interest[1,2] because of its anomalous behavior.[3-6] In a liquid heated at *constant density* we expect that the short-range order diminishes because of the increased thermal motion and in fact this is shown by the main peak of $S(k,T)$ that decreases in intensity and becomes wider with increasing temperature. In liquid helium the opposite behavior is observed as long as the temperature remains below T_λ, the temperature of the superfluid phase transition; the trend of $S(k,T)$ becomes normal above T_λ.

Two explanations have been proposed for the anomalous behavior or $S(k,T)$ below T_λ. The first[6] attributes this behavior of $S(k,T)$ to the hypothesized presence of Bose-Einstein condensation in the superfluid phase of ^{4}He. At a qualitative level it is said that He atoms in the condensate are delocalized and therefore do not contribute to spatial order. Since the condensate fraction $n_0(T)$ is a decreasing function of T that should vanish at and above T_λ, the anomalous behavior of $S(k,T)$ arises because more particles, $[1-n_0(T)]N$, contribute to give spatial order when the system is heated. This mechanism ends at T_λ and the normal behavior of $S(k,T)$ is reasonably expected thereafter.

The second explanation[7] attributes the anomaly to the thermal excitation of rotons. The structure factor $S(k,T)$ was computed starting from a model density matrix that incorporates the Landau picture of superfluid ^{4}He in terms of a gas of almost independent elementary excitations, phonons and rotons, and the Feynman picture of a roton as a density oscillation with a wave vector k_0 of order of the inverse of the interparticle distance. Phonons are more readily excited than rotons but they are long-wavelength disturbances and do not affect the short-range order to an appreciable extent. Above 1 K rotons become the dominant excitations and the thermal population of these states increases the probability of density fluctuations of wave vector $k \approx k_0$. Since $S(k,T) = \langle \rho_k \rho_{-k} \rangle_T / N$ where ρ_k is the density fluctuation operator of wave vector k, and k_0 is close to the position of the first maximum of $S(k,T)$ one concludes that the main peak of $S(k,T)$ increases its height with increasing T at least as long as rotons can be considered good elementary excitations. De Michelis, Masserini, and Reatto[7] (DMR) computed explicitly $S(k,T)$ from that density matrix and subsequently this result was found in good agreement with experiment.[5]

We have extended the computation of DMR in several respects. The model density matrix contains the ground-state wave function ψ_0 and DMR used a Jastrow wave function ψ_J to approximate ψ_0. However, within this approach it has been shown[8] that in order to compute $S(k,T)$ it is not necessary to know explicitly the complete wave function but only the ground-state radial distribution function $g_0(r)$ is needed because the thermal correlations are weak with respect to those due to ψ_0 so that a suitable perturbation scheme can be used. The availability of accurate experimental data[5] for $S(k,T)$ in ^{4}He at low temperature permits one to deduce $g_0(r)$ so that this perturbation scheme to compute $S(k,T)$ can be used for ^{4}He. This is an important step forward with respect to the computation of DMR because $S(k,T)$ can now be deduced using only other experimental data: the low-temperature $S(k,T)$ and the spectrum ϵ_k of elementary excitations. No assumption is needed on the form of the Hamiltonian and of ψ_0.

In view of the observed change in the temperature dependence of $S(k,T)$ at T_λ we have extended the computation of DMR at higher temperature by including in the density matrix the effect of the finite lifetime of the roton states in an empirical way. In this way we are able to describe the behavior of $S(k,T)$ even at T_λ or slightly above.

The content of the paper is the following. In Sec. II we discuss the model density matrix. In Sec. III our method to compute $S(k,T)$ is discussed and the results at several temperatures and densities are presented and compared to experiment. Finally Sec. IV contains a discussion of our results.

II. DENSITY MATRIX

There is good evidence[9] that the lowest excited states of liquid ^{4}He have the form

$$\psi_k(r_1 \cdots r_N) = F_k(r_1 \cdots r_N)\psi_0(r_1 \cdots r_N) \ , \tag{1}$$

where F_k is a function that carries a momentum $\hbar k$ and contains correlations only between a small number of particles. If F_k contains no correlation at all we have the Feynman[10] choice

$$F_k(r_1 \cdots r_N) = [NS_0(k)]^{-1/2}\rho_k \, , \ \rho_k = \sum_i \exp(i\vec{k}\cdot\vec{r}_i) \ , \tag{2}$$

whereas the Feynman-Cohen wave function[11] corresponds to assume that F_k contains correlations only between pairs. $S_0(k)$ is the ground-state structure factor and appears in F_k as a normalization constant. Penrose[12] has shown long ago how to construct the density matrix for an ensemble of such states (1) and (2) assuming that the excitations are independent so that the total excitation energy is $E_{\text{exc}} = \sum_k \epsilon_k N_k$. N_k is the number of excitations with wave vector k and ϵ_k is their energy. Penrose considered only the case of phonon excitations so that $\epsilon_k = \hbar ck$ and $S_0(k) = \hbar k/2mc$, where c is the sound velocity. His computation, however, does not require this specialization[13] and the full density matrix reads, in the coordinate representation

$$\langle r_1' \cdots r_N' |\rho| r_1 \cdots r_N\rangle = Q_N^{-1}\psi_0(r_1' \cdots r_N')\psi_0(r_1 \cdots r_N)\exp\left[-\tfrac{1}{2}\sum_{i,j}[h_1(r_i'-r_j')+h_1(r_i-r_j)+h_2(r_i'-r_j)]\right] \ , \tag{3}$$

where Q_N is the normalization constant determined by

$$\int dr_1 \cdots dr_N \langle r_1 \cdots r_N |\rho| r_1 \cdots r_N\rangle = 1 \ . \tag{4}$$

$h_1(r)$ and $h_2(r)$ are given by the Fourier integrals

$$h_1(r) = \frac{1}{(2\pi)^3\rho}\int d^3k \, \frac{1}{S_0(k)}\frac{\gamma_k^2}{1-\gamma_k^2} e^{i\vec{k}\cdot\vec{r}} \ , \tag{5}$$

$$h_2(r) = -\frac{1}{(2\pi)^3\rho}\int d^3k \, \frac{1}{S_0(k)}\frac{2\gamma_k}{1-\gamma_k^2} e^{i\vec{k}\cdot\vec{r}} \ . \tag{6}$$

ρ is the density and γ_k is the Boltzmann factor

$$\gamma_k = \exp(-\epsilon_k/k_B T) \ . \tag{7}$$

The only condition for the validity of the density matrix (3) when the complete spectrum ϵ_k is considered, is that the wave function corresponding to multiple excitations, for instance $F_k^{N_k}\psi_0/\sqrt{N_k!}$ when N_k excitations of wave vector k are present, is normalized for arbitrary k and this has been proved[9] under the separability hypothesis, i.e., the effects of the excitations of different wave vector[14] are additive. This is true when the number of excitations is finite and is a reasonable approximation when the average number of excitations is small compared with the number of particles.

The energy spectrum in Eq. (7) should be the one consistent with the wave function (1) and (2), i.e., it should be the Feynman spectrum

$$\epsilon_k = \hbar^2 k^2/2mS_0(k) \ , \tag{8}$$

but this is quantitatively unsatisfactory in the roton region and Feenberg[13] has justified the use in Eq. (7) of the empirical ϵ_k. If T_λ is approached closer than roughly 0.5 K experimentally it is observed that roton excitations have a temperature-dependent energy, $\epsilon_k(T)$ and acquire a finite lifetime. These are manifestations of roton-roton interaction and therefore are outside the description given by the density matrix (3). Feenberg[14] has already suggested the use of $\epsilon_k(T)$ in the density matrix[3] as a way to take into account part of the effects of the interaction. This will introduce more complicated correlations than the ones contained in Eq. (3) but one of its effects is simply to change the counting of the states by changing the average energy necessary to excite a roton.

We propose to make one further step in the inclusion of the interaction effects in the density matrix and precisely to take into account the finite lifetime of rotons. When the lifetime is infinite there is a one to one correspondence between the wave vector $\vec{k}$ of a density fluctuation in Eq. (2) and the energy in the Boltzmann factor (7). This correspondence is lost when the lifetime is finite and a density fluctuation $\rho_{\vec{k}}$ can correspond to many different energies centered around $\epsilon_k(T)$ with a dispersion $\delta_k(T)$ of order of the inverse lifetime. A reasonable assumption is that the spectral density is a Lorentzian and the linewidth $\delta_k(T)$ can be assumed to coincide with the linewidth measured by inelastic neutron scattering. Now each excitation of wave vector k does not carry the Boltzmann factor $\gamma_k = \exp(-\epsilon_k/k_BT)$ but the modified one

$$\bar{\gamma}_k = \frac{1}{m_k} \int_{\delta_k(T)}^{\infty} d\epsilon \frac{\delta_k(T)}{\delta_k^2(T) + [\epsilon - \epsilon_k(T)]^2} e^{-\epsilon/k_BT} \quad . \tag{9}$$

The density matrix turns out to have still the form (3) but in the expressions (5) and (6) for $h_1(r)$ and $h_2(r)$ $\bar{\gamma}_k$ replaces γ_k. In Eq. (9) the integral has the lower limit $\delta_k(T)$ because we assume that only excitations *not* overdamped ($\epsilon > \delta_k$) contribute to the coherent part of the density matrix. m_k is the normalization constant

$$m_k = \int_{\delta_k(T)}^{\infty} d\epsilon \frac{\delta_k(T)}{\delta_k^2(T) + [\epsilon - \epsilon_k(T)]^2} = \frac{1}{2}\pi - \arctan\left(1 - \frac{\epsilon_k(T)}{\delta_k(T)}\right) \quad . \tag{10}$$

In conclusion the density matrix (3) with γ_k given by Eq. (7) with the low-temperature excitation spectrum is a well-founded density matrix of liquid ^{4}He for temperature, at vapor pressure, up to ~ 1.7 K. Above this temperature interaction between rotons becomes important but the density matrix (3) still represents a useful model when it is used together with the empirical data for the energy $\epsilon_k(T)$ and linewidth $\delta_k(T)$ of roton states. In the range $1.8° < T < 2$ the lifetime of roton states is long enough ($\delta_k/\epsilon_k \lesssim 0.2$) that it can be neglected but above 2° the lifetime becomes short and the Boltzmann factor (7) should be replaced by $\bar{\gamma}_k$ given in Eq. (9). It should be noticed however that our treatment of the roton lifetime is only empirical and it lacks a microscopic justification. Our aim is principally to indicate the change of the behavior of $S(k,T)$ that takes place near T_λ. We have limited our considerations to the roton excitations as the most significant for short-range correlations. For long-wavelength phenomena phonon excitations should be considered.

The density matrix (3) does not take into account explicitly the effect of backflow[10] of the roton states. This problem is considered in Sec. IV.

III. TEMPERATURE DEPENDENCE OF CORRELATIONS

The radial distribution function is expressed in terms of the diagonal part of the density matrix

$$g(r_1 - r_2, T) = \frac{N(N-1)}{\rho^2} \int dr_3 \cdots dr_N \times \langle r_1 \ldots r_N | \rho | r_1 \ldots r_N \rangle \tag{11}$$

and on the basis of the model form (3) we can write

$$\langle r_1 \cdots r_N | \rho | r_1 \cdots r_N \rangle = Q_N^{-1} \psi_0^2(r_1 \cdots r_N) \times \exp\left(-\frac{1}{2} \sum_{i \neq j} \nu_T(r_i - r_j)\right) \quad , \tag{12}$$

where

$$\nu_T(r) = -\frac{2}{(2\pi)^3 \rho} \int d^3k \frac{1}{S_0(k)} \frac{\gamma_k}{1 + \gamma_k} e^{i\vec{k}\cdot\vec{r}} \tag{13}$$

and γ_k is the Boltzmann factor with the empirical energy spectrum $\epsilon_k(T)$

$$\gamma_k = \exp[-\epsilon_k(T)/k_BT] \tag{14}$$

if we do not take into account the finite lifetime of the excitations. If we take into account this finite lifetime in Eq. (13) γ_k should be replaced by $\bar{\gamma}_k$ given by Eq. (9). Instead of averaging γ_k as in Eq. (9) we have also considered the energy average over the Lorentzian distribution of the thermal factor, $-2\gamma_k/(1+\gamma_k)$, contained in ν_T. We find no significant difference between these two prescriptions because γ_k is small.

The computation of $g(r,T)$ from this density matrix is similar to the problem one encounters in the case of classical statistical mechanics, the Boltzmann factor for the potential energy $\exp[-\frac{1}{2}\sum_{i \neq j} v(r_i - r_j)/k_BT']$ replacing the diagonal part of the density matrix in Eq. (11). In particular $\nu_T(r)$ has the same role of an additional effective two-body potential that modifies the correlations contained in ψ_0^2 and methods borrowed from the theory of classical fluids can be used to compute $g(r,T)$. If ψ_0 is approximated by a Jastrow wave function

$$\psi_0 = \prod_{i<j} \exp[-\tfrac{1}{2}u(r_{ij})] \ ,$$

the Monte Carlo method can be used with $u(r) + \nu_T(r)$ having the role of $v(r)/k_B T'$ in the problem of classical fluids. $u(r)$ and $\nu_T(r)$, of course, are *a*-dimensional quantities and the similarity with the classical problem is with the interatomic potential divided by a temperature that we have called T'.

Such Monte Carlo computation has been performed by DMR[7] and for $u(r)$ the form was used as given by variational computation of the ground state of ^{4}He when the Lennard-Jones interatomic potential is assumed. In that computation $\nu_T(r)$ as given by Eq. (13) was used with the experimental $S_0(k)$ and $\epsilon(k,T)$.

The term $\nu_T(r)$ turns out to be weak compared with $u(r)$ and of somewhat longer range. This is exactly the situation handled by the perturbation theory of classical liquids[15]: the interatomic potential $v(r)$ is decomposed in a strong short-range part $v_0(r)$ and in a weak part $w(r)$ of longer range and $w(r)$ is treated on the basis of a suitable perturbation scheme. Therefore we can adopt this scheme in our quantum problem if we identify the ground-state Jastrow correlation function $u(r)$ with the "reference" potential $v_0(r)/k_B T'$ of the classical system and the temperature-dependent term $\nu_T(r)$ with the perturbation term $w(r)/k_B T'$. In a previous work[8] (hereafter called I) we have followed this approach and we have computed $g(r,T)$ with the use of the so-called "exp" approximation of Andersen and Chandler.[16] $g(r,T)$ was computed also by the Monte Carlo method and from the good agreement between the two computations it was concluded that the "exp" approximation is very good indeed in this problem.

$g(r,T)$ in the exp approximation reads[8,16]

$$g(r,T) = g_0(r)\exp C_T(r) \ , \tag{15}$$

where $g_0(r)$ is the radial distribution function at $T = 0°$ and $C_T(r)$ is given by

$$C_T(r) = -(2\pi)^{-3}\int d^3k\, e^{-i\vec{k}\cdot\vec{r}}\frac{S_0^2(k)\nu_T^*(k)}{1+\rho S_0(k)\nu_T^*(k)} \ . \tag{16}$$

$S_0(k)$ is the structure factor corresponding to $g_0(r)$:

$$S_0(k) = 1 + \rho\int d^3r\, e^{i\vec{k}\cdot\vec{r}}[g_0(r)-1] \ , \tag{17}$$

and $\nu_T^*(k)$ is the Fourier transform of the "optimized" perturbation $\nu_T^*(r)$

$$\nu_T^*(k) = \int d^3r\, e^{i\vec{k}\cdot\vec{r}}\nu_T^*(r) \ . \tag{18}$$

This "optimized" perturbation is defined as follows:

$$\nu_T^*(r) = \begin{cases} \nu_T(r) & \text{for } r > d \\ \nu_T^0(r) & \text{for } r < d \ , \end{cases} \tag{19}$$

where $\nu_T^0(r)$ is such that $C_T(r) = 0$ for $r < d$, and d represents the diameter of the equivalent hard-sphere system.[17] In the theory of classical fluids the motivation for using this optimized potential is the following[16]: the strong repulsive part of the interatomic potential at short distance renders in practice inaccessible those configurations in which at least one couple of particles has the relative distance $|\vec{r}_i - \vec{r}_j|$ less than a distance d that can be considered as the diameter of an equivalent hard-sphere system. This implies that any measurable quantity, in particular $g(r)$, does not depend on the form of the perturbation $w(r)$, that in our case is $\nu_T(r)$, for $r < d$, i.e., the shape of $w(r)$ for $r < d$ is arbitrary. Thus one takes advantage from the arbitrariness of $w(r)$ for $r < d$ by choosing that particular form $w^*(r)$ for which the perturbation series is the most rapidly convergent and this gives[16] the condition $C_T(r) = 0$ for $r < d$. The same can be said in the quantum case: because of the strong repulsive part of the He-He interatomic potential ψ_0 vanishes rapidly whenever $|\vec{r}_i - \vec{r}_j| < d$ and the concept of an equivalent quantum hard-sphere system is a good approximation[17] (if one uses the Lennard-Jones potential to represent the He-He interaction, it turns out that $d \approx 0.81\sigma$, $\sigma = 2.556$ Å). Thus the use of Eq. (19) as optimized perturbation is justified and in I we have verified the good accuracy of this approximation. Moreover we have also verified that $g(r,T)$ is not affected by small variations of the equivalent hard-sphere diameter d around the value 0.81σ.[18]

The possibility of using the perturbation scheme to compute $g(r,T)$ is important because it gives the possibility of computing $g(r,T)$ by using only experimental data. In fact within the perturbation scheme [Eqs. (15) and (16)] one needs to know of the ground state ψ_0 only $g_0(r)$ and $S_0(k)$ and these quantities can be deduced from accurate neutron or x-ray scattering experiments performed at low temperature as the one recently completed.[5] Of course the exact ground state does not have the simple Jastrow form but the "exact" computation[19] of the ground state of the quantum Lennard-Jones fluid with the Green's-function Monte Carlo method confirms that the dominant correlations in ψ_0 are well represented by the Jastrow form ψ_J. The most important correlations absent from ψ_J are three-particle correlations[20,21] but also for these one can develop an analogy with a problem in classical statistical mechanics of a fluid with three-body interatomic interaction in addition to the two-body one. For the purpose of computing $g(r)$ it has been shown[22] that a three-body interaction can be replaced by an effective two-body potential and thus the analysis leading to [Eqs. (15) and (16)] can be repeated. We conclude that we are justified in using those expressions together with the experimental g_0 and S_0 to evaluate the temperature effect on correlations.

In our computation at the equilibrium density[23] $\rho_{eq} = 0.145\ \mathrm{g\,cm^{-3}}$ for $S_0(k)$ we have used the $S(k)$ of Svennson *et al.*[5] at $T = 1$ K modified at small k so that they are consistent with the $\hbar k/2mc$ behavior characteristic of the ground state. $g_0(r)$ has been obtained from this $S_0(k)$ by Fourier inversion of Eq. (17). The temperature-dependent correlation term $\nu_T(r)$ has been computed using in Eq. (14) the experimental spectrum $\epsilon_k(T)$. Only few complete spectra have been measured in ^{4}He and for this reason we have considered only the roton contribution, that in any case is the most important for our purpose, with the energy spectrum represented by a parabola:

$$\epsilon_k(T) = \Delta(T) + \hbar^2[k - k_0(T)]^2/2\mu(T) \ . \tag{20}$$

This approximation is adequate as can be seen in Fig. 1 where we compare the temperature effect on the structure factor

$$\Delta S(k,T) = S(k,T) - S_0(k) \tag{21}$$

computed with the full spectrum[24] at 2.1° and with the parabola (20) with the value of the parameters ($\Delta = 7.33°$, $k_0 = 1.91$ Å^{-1}, and $\mu = 0.13 m_{\mathrm{He}}$) fitted on the same data. In the region of the main maximum of $S(k)$ there is good agreement between the two results and all other computations on which we report have been obtained with the analytic representation (20) of $\epsilon_k(T)$.

In agreement with the result of DMR obtained from a Monte Carlo computation of $g(r,T)$, the main temperature effect on the structure factor is an increase and a sharpening of the main maximum of $S_0(k)$ and this corresponds to the characteristic behavior of $\Delta S(k,T)$ positive at the peak of S_0 followed by two negative minima at both sides. There is also a rather small displacement (~ 0.1 Å^{-1}) to lower k of the position of the maximum of ΔS compared with that of S_0. The difference between our present result and the one of DMR is not due to the different method used to compute $g(r,T)$ but to our use of the experimental $g_0(r)$ instead of using a variational ground-state wave function. In addition now we have computed the temperature correlation $\nu_T(r)$ using the most recent data[5] for $S_0(k)$.

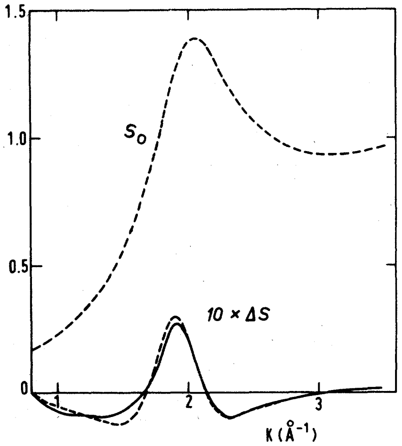

FIG. 1. Temperature effect, $\Delta S(k,T) = S(k,T) - S_0(k)$, at ρ_{eq} and $T = 2.1°$ with the use of the full spectrum (solid line) and of the roton parabola (20) (dashed line). $S_0(k)$ is also shown.

In order to compare our results with the experimental data[5] we have computed $S(k,T)$ at the same temperatures of the experiments. For the parameters of the roton spectrum (20) we have used the experimental data[25,26] that have been smoothed and interpolated. The values we have used are listed in the Appendix. We have also computed $S(k,T)$ taking into account the finite lifetime of rotons by using Eq. (9). Below T_λ the linewidth $\delta_k(T)$ is almost proportional to the energy $\epsilon_k(T)$ and good representation of δ_k is given by[25]

$$\delta_k(T) = [\Gamma + G(k - k_0)]\epsilon_k(T) \ , \tag{22}$$

where Γ and G are two constants that depend on temperature. The slope G however is rather small and we have neglected it also in view of the approximate representation of $\epsilon_k(T)$ by the parabola outside the region of the minimum. In Fig. 2 our result for $S(k,T)$ at $T = 2.15°$ is shown when we take into account the finite lifetime and when we consider sharp excitations. The shape of $\Delta S(k,T)$ is not affected by the finite lifetime of rotons but the amplitude of the temperature effect is reduced and this effect becomes more significant with increasing temperature because $\Gamma = \delta_{k_0}/\epsilon_{k_0}$ increases very rapidly when $T \sim T_\lambda$. In Fig. 2 also the experimental data[5] are shown [we report the unsmoothed differences $S(k,T) - S(k,T = 1°)$]. There is an overall good agreement between theory and experiment in the shape and in the amplitude of $\Delta S(k,T)$, the main difference consisting in a

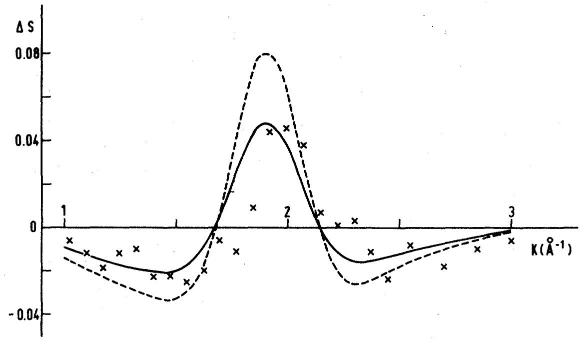

FIG. 2. $\Delta S(k,T)$ at ρ_{eq} and $T = 2.15°$. Experimental data (Ref. 5) (crosses) and theory for finite linewidth (solid line) and for sharp excitation ($\delta_k = 0$) (dashed line).

small (~ 0.1 Å^{-1}) displacement of the theoretical curve compared to the experimental one. In fact experiment shows almost no displacement with T of the position of the maximum of $S(k,T)$. On the contrary our result shows a systematic even if small displacement of the maximum of $S(k,T)$ compared with that of $S_0(k)$ to a smaller wave vector. For instance at $T=2.15°$ the maximum of $S(k,T)$ is found at $k \approx 2.025$ Å^{-1} in place of $k \approx 2.04$ Å^{-1} for $S_0(k)$. The origin of this displacement can be easily understood. The maximum of $\Delta S(k,T)$ is for that k for which the density fluctuation is most easily thermally excited and for our model density matrix this corresponds roughly to the position k_0 of the roton minimum because we have adopted the Feynman wave function. The roton minimum is at a smaller wave vector than the maximum of $S(k)$, the displacement is roughly ~ 0.1 Å^{-1}, and we believe that this is responsible in our theory for the displacement of the maximum of $S(k,T)$. As we discuss in Sec. IV the agreement with experiment should improve if one takes into account the backflow associated with roton excitations.

The shape of $\Delta S(k,T)$ reported in Fig. 2 is typical also for the other temperatures (see Fig. 4 below for the result at $T=1.97°$) and we report only the maximum value of the difference $\Delta S(k,T)$ as function of T in Fig. 3. When we do not take into account the finite lifetime of rotons the maximum of $S(k,T)$ steadily increases with T because rotons become more easily excited for the direct effect of the increased thermal energy and also because the roton energy $\Delta(T)$ decreases with increasing T. When we take into account the finite lifetime of rotons the temperature effect on $S(k,T)$ is strongly reduced for $T \approx T_\lambda$ because the linewidth becomes comparable with ϵ_k and as a consequence the temperature effect is no more concentrated on density fluctuations of a well-defined k value. This effect is so important that the increase of the maximum of $S(k,T)$ with temperature ends slightly above T_λ where $\delta_{k_0} > \epsilon_{k_0}$, in good agreement with experiment. We recall that the theory does not contain any adjustable parameter. We find that the maximum value in the temperature effect on $S(k,T)$ is about 20% below the experimental value but we believe that this difference is not significant. Experiments are not accurate enough to permit a very precise determination of these small differences on the structure factor. On the other hand the theoretical results are affected by the precision of the data available on $\epsilon_k(T)$. In fact the scatter in the values[24] of the roton parameters of the parabola (20) given by experiment is rather large particularly around T_λ. We believe more important, however, that when $T \gtrsim T_\lambda$ the linear relation (22) between δ_k and ϵ_k does not give a good representation of experiment because δ_k/ϵ_k has a maximum[24] at $k \approx k_0$. Unfortunately the published data are not complete enough to allow for this effect in our computation and only the value of this maximum $\Gamma = \delta_{k_0}/\epsilon_{k_0}$ is available. As a consequence our computation overestimates the effect of the finite lifetime of rotons with k in the neighborhood of k_0 when $T \gtrsim T_\lambda$. To give an estimate of this effect we have performed a computation at two temperatures ($T=2.20°$ and $2.27°$) decreasing the experimental value of Γ by 20%, a change that is reasonable in view of the available data.[25] As it can be seen in Fig. 3 our result is rather sensitive to the value of Γ and we conclude that theory and experiment agree within their reciprocal uncertainty. We are not able to compare our result with the x-ray data of Robkoff *et al.*[1] because they do not report the complete $S(k)$ and their data do not extend to low enough temperature.

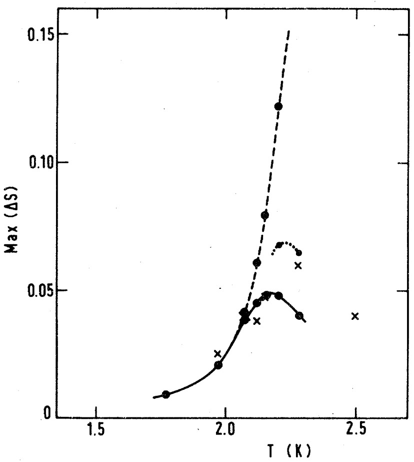

FIG. 3. Maximum value of $\Delta S(k,T) = S(k,T) - S_0(k)$ as function of temperature at ρ_{eq}. Experimental data (Ref. 5) (crosses) and theory for finite linewidth (dots connected by a solid line), for reduced linewidth 0.8Γ (dotted line) and for sharp excitations (dots connected by a dashed line).

If in the computation of $S(k,T)$ one makes the approximation of separability of the effect of excitations of different wave vector the result will be the simple formula[13]

$$S(k,T) = S_0(k) \coth[\epsilon_k(T)/2k_B T] \ . \tag{23}$$

This approximation coincides with the so-called RPA (random phase approximation) in the theory of classical fluids and it corresponds to

$$g(r,T) = g_0(r) + C_T(r) \tag{24}$$

in place of Eq. (15) with $C_T(r)$ given by Eq. (15) but *without* "optimization" of the perturbation $\nu_T(r)$. The approximation (23) is not satisfactory as it can be seen in Fig. 4 because it overestimates the increase of the maximum of $S(k,T)$ and $\Delta S(k,T)$

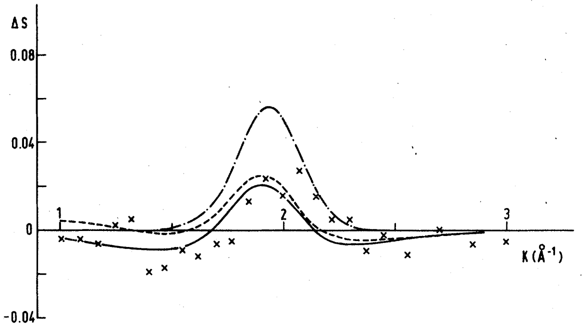

FIG. 4. $\Delta S(k,T)$ at ρ_{eq} and $T=1.97°$. Experimental data (Ref. 5) (crosses) and theory for sharp excitations with the "exp" approximation (15) (solid line), with the linear approximation (24) with "optimized" perturbation (19) (dashed line) and with Feenberg formula (23) that corresponds to Eq. (24) used with the bare perturbation $\nu_T(r)$ (dotted-dashed line).

$=S_0(k)[\coth(\epsilon_k/2k_BT)-1]$ is always positive so that there is no sharpening of the maximum. In order to understand the origin of the failure of the separability hypothesis we have also computed $S(k,T)$ on the basis of the linear relation (24) but we have used for $C_T(r)$ the "optimized" perturbation $\nu_T(r)$ as given by Eq. (19). From the comparison of the different results reported in Fig. 4 we conclude that the failure of (23) is due primarily to the absence of the optimization condition. When one computes a correlation function the effect of exciting density fluctuations of different wave vectors is not simply additive because the necessity of satisfying the core condition [$g(r)=0$ at small distance] forces a correlation between the different wave vectors and this is exactly the role of the optimization condition.

The temperature dependence of $S(k,T)$ is enhanced at higher densities. We have computed $S(k,T)$ along two isocores, at $\rho=1.1\times\rho_{eq}=0.1595$ g cm^{-3} and at $\rho=1.2\times\rho_{eq}=0.174$ g cm^{-3}. This corresponds roughly to $p\approx 10$ and 24 atm. Above the equilibrium density accurate enough data for $S(k)$ are not yet available and we have used for $g_0(r)$ and $S_0(k)$ the results[19] of the Green's-function Monte Carlo method for the Lennard-Jones potential. The smoothed roton energies and lifetimes obtained from literature[25] are reported in the Appendix. The behavior of $S(k,T)$ is very similar to the one found at equilibrium density (see Fig. 5): the temperature effect on the maximum of $S(k,T)$ reaches a maximum slightly above T_λ and there is a small displacement of the maximum of $S(k,T)$ to smaller wave vector. The height of the maximum of $S(k,T)$ increases significantly at higher densities because the more pronounced short-range order present in the ground state at high density makes it easier to excite additional density fluctuations [cf. the presence of $S_0^{-1}(k)$ in Eq. (13) for $\nu_T(r)$] and the effective mass μ of rotons decreases so that the temperature effect is more concentrated around k_0. The decrease in the roton energy Δ at high density is responsible only for the overall displacement of the curve at lower temperature because the ratio Δ/T_λ is roughly independent of ρ. Experimental data are not yet available for a detailed comparison with our result. The only measurement, in fact, along different isocores is the x-ray data of Robkoff *et al.*[1] but unfortunately they did not measure $S(k)$ to low enough temperature to allow a determination of $S_0(k)$. In any case their data are consistent with an increase of the temperature dependence of $S(k,T)$ at high density and the difference is of the same order as the one we find. For the maximum of $S(k,T)$ they find $[S_{max}(2.1°)-S_{max}(1.8°)]/S_{max}(1.8°)=0.041$ at the higher density $\rho=0.1625$ g cm^{-3} against $[S_{max}(2.2°)-S_{max}(1.9°)]/S_{max}(1.9°)=0.031$ at the lower density $\rho=0.1503$ g cm^{-3}.

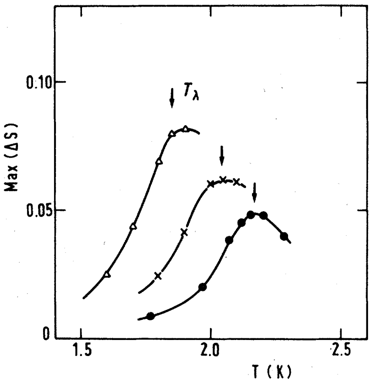

FIG. 5. Maximum value of $\Delta S(k,T)$ as function of temperature at three densities: ρ_{eq} (dots), $1.1\times\rho_{eq}$ (crosses), and $1.2\times\rho_{eq}$ (triangles). The corresponding temperatures of the λ transition are shown by the arrows.

A careful determination of the temperature dependence of $S(k,T)$ at different densities is important[27] because it can discriminate between the present theory and the interpretation of the T dependence of $S(k,T)$ in term of Bose-Einstein condensation. In this last case, in fact, the T dependence of $S(k,T)$ should decrease, and not increase, as one approaches solidification because the condensate decreases for increasing density.

IV. DISCUSSION

The main features of the temperature dependence of the structure factor $S(k,T)$ of liquid ^{4}He are well explained in terms of thermal population and renormalization of roton states. We have shown in fact that Penrose's density matrix constructed with the empirical temperature-dependent spectrum and linewidth of roton states give $S(k,T)$ in good agreement with experiment. On this basis we get a clear picture of the origin of the anomalous behavior of ^{4}He. In a fluid where classical mechanics is a good approximation the static correlations are decoupled from the dynamics and in fact $g(r)$ does not depend at all on the mass of the atoms. In this case the temperature dependence of correlations is due only to the trivial T dependence of the Boltzmann factor for the potential energy and from this the decrease of the short-range order with increasing T follows directly. In a quantum system, instead, static correlations and dynamics are strictly related. In a Bose fluid like ^{4}He the presence of short-range order is at the basis of the existence of roton excitations, i.e., of excitations of low energy at a nonzero wave vector that roughly corresponds to the wave vector of the short-range order. Thus the temperature dependence of the properties of the fluid, and in particular of $g(r)$, are determined by these excitations. By increasing T more rotons are present but this corresponds to an enhancement of short-wavelength density fluctuations and therefore of the short-range order. This trend continues as long as rotons are well-defined elementary excitations. When the lifetime becomes too short the correspondence of energy to wave vector is lost and an increase of temperature does not preferentially enhance density fluctuations of a well-defined wave vector. Thus the behavior of $S(k,T)$ found in ^{4}He is expected to hold in any high-density Bose fluid. The strict relation between correlations and dynamics has already been noticed[8,20] in the ground state of strongly interacting Bose fluid.

An interesting question is if this anomalous behavior is present also in a Fermi liquid, for instance in liquid ^{3}He. The results for a Bose fluid cannot be transposed to this case because a short-wavelength density fluctuation does not represent an elementary excitation in a Fermi system because the continuum of particle-hole excitations gives a channel for decay. However inelastic neutron scattering[28] shows that the average energy transfer as function of k has a modulation reminiscent of the roton spectrum in ^{4}He. To be precise the inelastic peak is quite wide but still the modulation is evident and one can wonder if also in ^{3}He $S(k,T)$ has an anomalous behavior. At saturated vapor pressure (SVP) the linewidth is so large that we do not expect that $S(k,T)$ of ^{3}He has the anomalous behavior of ^{4}He but this might appear at high pressure near solidification where the peaks become narrower. An experimental determination of the temperature dependence of $S(k,T)$ in ^{3}He would be very interesting.

Our computations are based on a model density matrix that is based on certain assumptions that we now discuss. At the basis of the density matrix is the separability hypothesis that allows the construction of the wave functions with many excitations simply in term of the single excitation ones. The hypothesis is correct in the long-wavelength limit but for arbitrary k it is rigorously true only when the number of excitations is finite.[9] In the density matrix the average number of excitations is of order of the number of particles and separability is no more rigorous. Justifications for the use of this hypothesis even in this case have been given[9,13] but some doubt can arise because we have shown in Sec. III that this hypothesis leads to inaccurate result when it is used to compute $S(k,T)$. We notice however that this hypothesis enters in a different way when we compute $S(k,T)$ or when we construct the density matrix. In the computation of $S(k,T)$ we have shown that separability fails because it leads to violation of the core condition that $g(r)=0$ at short distance. In the construction of the density matrix separability is used[12] in order to express the normalization constants that involve averages of type $\langle \psi_0 | |\rho_k|^{N_k} |\rho_{k'}|^{N_{k'}} | \psi_0 \rangle$ in term of product of the simple average $\langle \psi_0 | |\rho_k|^2 | \psi_0 \rangle = N S_0(k)$. By using the exact $S_0(k)$ the core condition is automatically satisfied and the result of Sec. III does not object to the use of separability for the density matrix.

If we try to describe ^{4}He near the λ point it is necessary to take into account the finite lifetime of rotons. We have included this effect in an empirical, but we believe reasonable, way. We have assumed that in averaging the density matrix over the distribution of the energy of the excitations we can treat each excitation independently of the others and this is consistent with our picture of elementary excitations. More questionable is the introduction of the lower limit δ_k in the average (9) of the Boltzmann factor. The assumption here is that overdamped ($\epsilon < \delta_k$) excitations cannot be treated as the underdamped ones on the basis of a wave function of the form of Eqs. (1) and (2) and that these overdamped excitations give some different kind of contribution to the density matrix that is spread out more uniformly in k space so that it can be neglected. In any case our results do not depend significantly on the exact value of this lower limit. In fact we have also performed computations of $S(k,T)$ when the average (9) is over the interval $(0-\infty)$ and the results are quite similar to the ones presented in Sec. III. Our model density matrix gives always a finite Bose-Einstein condensate n_0 even when we apply it to temperature $T > T_\lambda$. This is clearly an unsatisfactory aspect of our density matrix but we do not believe that it is very significant for our purpose: n_0 is related to the off-diagonal element of the density matrix when $|r_i - r_j| \rightarrow \infty$ whereas $g(r,T)$ is determined by the diagonal part when couples of particles are at microscopic mutual distances.

The accuracy of our computation is limited by the limited amount and by the scatter of the data on the spectrum $\epsilon_k(T)$ of the roton states. On the other hand we do not believe that one can ask from our model density matrix more than a semiquantitative agreement with experiment because at not too high temperature ($T\sim1.5°$) where our density matrix is well founded the difference $S(k,T)-S_0(k)$ is just too small to be measured reliably whereas at higher T it is our density matrix that becomes approximate. However experiment and theory differs in one significant feature. Our theory gives a systematic displacement, not present in the experimental data, in the position of the main maximum of $S(k,T)$ to a lower k value when the temperature is increased. We have already discussed in Sec. III that this is due to the different position in k space of the minimum of ϵ_k and of the maximum of $S_0(k)$. We attribute this discrepancy to the use of the Feynman form [Eqs. (1) and (2)] for the wave function of the excitations. It is known that a roton is not simply a density fluctuation but important correlations are present in the form of backflow.[11] The excitation spectrum $\epsilon_k=\hbar^2k^2/2mS_0(k)$ computed with the Feynman wave function has the roton minimum roughly at the position of the maximum of $S_0(k)$. When the backflow is included the roton minimum is displaced to a smaller wave vector in agreement with experiment. Therefore it is conceivable that if the effects of the backflow were included in the density matrix this would modify the position in k space where the temperature effect is more effective. The effect of the backflow on the ground state has been considered in I and a similar argument can be extended at finite temperature. One finds that the backflow associated to rotons modifies the two-body term (13) but in addition introduces three-body correlations that are temperature dependent. Also in this case methods borrowed from the theory of classical liquids could be used to compute $S(k,T)$.

The anomalous temperature dependence of $S(k,T)$ has also been interpreted[1,2,6] as a manifestation of the T dependence of the Bose-Einstein condensation. The theoretical foundation of this interpretation however, has been shown[27] to be very feeble; in any case since our density matrix does have a finite Bose-Einstein (BE) condensate, it might be asked if the Cummings *et al.* formula[6] is in some way related to our theory. We do not believe that this is the case for the following two reasons. For a two-dimensional system our density matrix does not have any BE condensate[12] but still $S(k,T)$ has an anomalous temperature behavior as in three dimensions as long as rotons are well defined. No anomaly is predicted on the basis of the Cummings *et al.* formula. Second our theory predicts that the change in the height of

TABLE I. Parameters of the roton spectrum (20) and reduced linewidth Γ used in our computations.

T (K)	Δ (K)	μ/m_{He}	Γ
$\rho=\rho_{\text{eq}}$			
1.77	8.32	0.154	0.02
1.97	7.62	0.144	0.20
2.07	6.57	0.132	0.36
2.12	5.81	0.126	0.50
2.15	5.27	0.123	0.62
2.20	4.43	0.117	0.85
2.27	3.32	0.109	1.35
$\rho=1.1\times\rho_{\text{eq}}$			
1.80	6.68	0.135	0.21
1.90	5.86	0.130	0.36
2.00	4.72	0.122	0.58
2.05	3.93	0.120	0.79
2.10	3.02	0.116	1.13
$\rho=1.2\times\rho_{\text{eq}}$			
1.60	5.81	0.120	0.23
1.70	5.09	0.120	0.35
1.80	3.65	0.120	0.61
1.85	2.85	0.120	0.84
1.90	1.88	0.120	1.41

the main maximum of $S(k,T)$ between T_λ and low temperature *increases* strongly when the density is changed from the equilibrium value to the solidification one. On the contrary on the basis of the Cummings *et al.* formula a strong *decrease* is expected under the same conditions because all theories predict a decrease of the value of the BE condensate when the density is increased and an "exact" computation[19] has given a reduction of the BE condensate for more than a factor of 2 when the density is changed from the equilibrium value to the solidification one. An experimental verification of the temperature dependence of $S(k,T)$ along different isocores will be able to discriminate between these two different theories.

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APPENDIX

Parameters of the roton spectrum (20) and reduced linewidth $\Gamma = \delta_{k_0}/\epsilon_{k_0}$ at different temperatures and densities. The position of the roton minimum k_0 is kept constant at different T with the values $k_0 = 1.915$, 1.98, and 2.025 Å at density $\rho = \rho_{\mathrm{eq}}$, $1.1 \times \rho_{\mathrm{eq}}$, and $1.2 \times \rho_{\mathrm{eq}}$, respectively. The values of the parameters have been obtained from interpolation of the experimental data (Refs. 25 and 26). (See Table I.)

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