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Two-particle excitations in liquid He II

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It is suggested that two-particle excitations in liquid He II may be yet another source for the observed loss in spatial order in liquid ^{4}He as it is cooled through T_λ.

Recent measurements of the structure factor for liquid ^{4}He at constant density above and below T_λ by Robkoff *et al.*[1] have revealed that the maximum height of the main structure-factor peak increases as the temperature is lowered towards T_λ, but suddenly starts decreasing as the temperature is still lowered below T_λ, indicating a loss in spatial order on cooling through T_λ. Theoretically it has been suggested that the observed loss in spatial order may either be due to Bose-Einstein condensation[2] or due to the thermal excitation of rotons.[3] Recently a number of critical studies of the suggestion of Hyland *et al.*[2] have resulted in uncertainty about the suggestion that the observed loss in spatial order is due to Bose-Einstein condensation.[4–6] While the mechanism of thermal excitation of rotons seems plausible, it seems worthwhile to investigate a third possible explanation—the production of two-particle excitations. The aim of the present investigation is to stress the third possibility.

Since the neutron scattering experiment of Cowley and Woods,[7] which showed the presence of a second diffuse branch above the well-known phonon-roton branch, a number of theoretical investigations have been carried out to interpret the new branch as due to two-particle excitations. On employing the Tamm-Dancoff approximation (i.e., by restricting the intermediate states to one- and two-particle states only), Iwamoto[8] obtained the second branch as a resonance of particle-particle scattering. Kebukawa *et al.*[9] obtained the upper branch as a second solution of their equation for the excitation energy obtained on the basis of a collective variable theory. Enz[10] obtained the new branch corresponding to the second eigenvalue of the equation of motion for the density response function. Extension of a theory due to Vasudevan *et al.*[11] based on currents and densities by Sridhar[12] yields a second branch that is almost constant for small values of momentum. Khalatnikov[13] also interprets the data of Cowley and Woods as indicating a second branch due to two-particle excitations. Fukushima *et al.*[14] also obtain a similar conclusion.

Raman scattering experiments on liquid He II performed by Greytak and Yan[15] also seem to confirm the possible existence of a second branch. According to Ruvalds *et al.* and Zawadowski *et al.*,[16] who have considered the effect of roton-roton interactions on the scattering cross section, a bound state of two rotons with very small relative momentum will explain the frequency and intensity of the observed peak in the Raman scattering experiment.

There is a view that the new branch is actually a band and does not correspond to a well-defined frequency. However, Soda *et al.*[17] have satisfactorily explained the experimental intensity curves by assuming that the second branch corresponds to a well-defined frequency. The intensity function for the deep inelastic scattering of neutrons has been satisfactorily obtained by Wong[18] by approximating the multiphonon background of the dynamic-structure function with two-particle excitations only. A similar attempt has been made by Tripathi *et al.*[19]

Interest in two-particle excitations has recently been revived because of the following developments:

(a) Cowley[20] doubted whether the Raman scattering experiments decisively establish the existence of a two-particle bound state while Kleban[21] arrives at a similar conclusion from a theoretical point of view.

(b) Recent neutron scattering data obtained by Blagoveshchenskii *et al.*[22] indicate that there may arise a second branch due to a two-roton bound state.

These developments indicate the necessity of investigating the two-particle excitations in a greater depth.

Miller *et al.*[23] have suggested that scattering at low temperatures may be divided into one-phonon and multiphonon parts in the following manner:

$$S(\vec{k},\omega)=S_F(k)\delta(\omega-\omega_1(k))+S_{II}(\vec{k},\omega)\ , \qquad (1)$$

where $S_F(k)$ gives the strength and $\omega_1(k)$ gives the energy of the one-phonon excitation, while $S_{II}(\vec{k},\omega)$ gives the multiphonon background.

The aim is to specify $S_{II}(\vec{k},\omega)$ under the assumption that single- and two-particle excitations saturate the sum rules and that these excitations have fairly long lifetimes. To estimate an approximate form for $S_{II}(\vec{k},\omega)$ one can use the form of $S(\vec{k},\omega)$ are derived by Soda *et al.* [Eq. (21) of Ref. 24] in the limit where the width of the excitations Γ becomes very small. With a little algebra one can show that the expression of Soda *et al.*[24] in the limit of vanishing width is equivalent to

$$S(\vec{k},\omega)=\frac{\langle 0|\rho^{\dagger}(\vec{k})\rho(\vec{k})|0\rangle}{|\omega_1-\omega_2|}\langle\Delta\omega^2\rangle_k \times\left[\frac{\delta(\omega-\omega_1)}{\omega_1-\langle H\rangle_k}+\frac{\delta(\omega-\omega_2)}{\omega_2-\langle H\rangle_k}\right]\ , \qquad (2)$$

where $\omega_1=\omega_1(k)$ and $\omega_2=\omega_2(k)$ represent the single-particle and two-particle energies when the sum rules are assumed to be saturated with single- and two-particle excitations only. $\langle\Delta\omega^2\rangle_k$ represents the mean-square fluctuation in energy about the mean-excitation energy $\langle H\rangle_k=[k^2/2mS(k)]$, $S(k)$ being the static structure factor. Comparing (2) and (1) one can write approximately

$$S_{II}(\vec{k},\omega)=S_{II}(k)\delta(\omega-\omega_2(k))\ . \qquad (3)$$

An approximate form for $\omega_2(k)$ is derived from the following considerations:

(i) Iwamoto[8] obtains a second branch which is somewhat flat with energy 20 K up to a wave number 2.3 Å^{-1}.

(ii) The equation of motion for the density response function as evaluated by Enz[10] leads to a second mode, which for large values of the momentum exhibits free-particle behavior while for small momenta has an energy approximately equal to twice the roton gap.

(iii) Soda *et al.*[17] also obtained a similar behavior for $\omega_2(k)$.

(iv) Sridhar[12] also obtained a second branch that is almost constant for small momentum values.

Consequently, the following form for $\omega_2(k)$ is assumed. This form gives a constant value for the second branch for small momentum values and yields free-particle behavior for large values of k:

$$\omega_2(k)=\begin{cases}a\ \mathrm{K}\ \ \text{for } k\le k_0\\ (\hbar^2k^2/2mk_B)\ \mathrm{K}\ \ \text{for } k\ge k_0\end{cases} \qquad (4)$$

where a and k_0 are constants chosen such that

$$\omega_2(k_0)=a=\hbar^2k_0^2/2mk_B\ . \qquad (5)$$

From the experimental data (Fig. 6 of Ref. 7) a can be varied over the range

$$20\ \mathrm{K}\le a\le 25\ \mathrm{K}\ .$$

In the above k_B denotes the Boltzmann constant. The momentum transfers in the above forms are less than 2.5 Å^{-1}. Following standard procedure the first few moments of $S(\vec{k},\omega)$ can be computed easily:

$$\frac{1}{2\pi}\int_0^\infty d\omega S(\vec{k},\omega)=\langle 0|\rho(\vec{k})\rho^{\dagger}(\vec{k})|0\rangle=NS(k)\ , \qquad (6)$$

$$\frac{1}{2\pi}\int_0^\infty d\omega\,\omega S(\vec{k},\omega)=\langle 0|\rho(\vec{k})H\rho^{\dagger}(k)|0\rangle=NE_0(k)\ , \qquad (7)$$

$$\frac{1}{2\pi}\int_0^\infty d\omega\,\omega^3 S(\vec{k},\omega)=\langle 0|\rho(\vec{k})H^3\rho^{\dagger}(\vec{k})|0\rangle=N[E_0(k)\omega_1^2(k)+F(k)]\ , \qquad (8)$$

where

$$E_0(k)=\hbar^2k^2/2m\ , \qquad (9)$$

$$\omega_1(k)=[E_0^2(k)+2NE_0(k)V(k)]^{1/2}\ , \qquad (10)$$

$$F(k)=4E_0^2(k)\langle\mathscr{E}_k\rangle+\frac{\hbar^4}{2m^2}\sum_{\vec{p}}\{V(|\vec{p}+\vec{k}|)[\vec{k}\cdot(\vec{p}+\vec{k})]^2-V(p)(\vec{k}\cdot\vec{p})^2\}[S(p)-1]\ . \qquad (11)$$

In the above $V(k)$ is the two-particle interaction potential and $\langle\mathscr{E}_k\rangle$ is the average kinetic energy. $\omega_1(k)$ approximates the one-phonon energy for large k while it is exact for values of $k\le 0.6$ Å^{-1}. Equations

(6)–(8) can be written as

$$NS(k)=\frac{|\langle 0|\rho(\vec{k})|1\rangle|^2}{\langle 1|1\rangle}+\sum_{n\neq 1}\frac{|\langle 0|\rho(\vec{k})|n\rangle|^2}{\langle n|n\rangle}, \tag{12}$$

$$NE_0(k)=\omega_1(k)\frac{|\langle 0|\rho(\vec{k})|1\rangle|^2}{\langle 1|1\rangle}+\sum_{n\neq 1}\omega_n(k)\frac{|\langle 0|\rho(\vec{k})|n\rangle|^2}{\langle n|n\rangle}, \tag{13}$$

$$N[E_0(k)\omega_1^2(k)+F(k)]=\omega_1^3(k)\frac{|\langle 0|\rho(k)|1\rangle|^2}{\langle 1|1\rangle}+\sum_{n\neq 1}\omega_n^3(k)\frac{|\langle 0|\rho(k)|n\rangle|^2}{\langle n|n\rangle}. \tag{14}$$

The state $|1\rangle$ is the one-phonon state which saturates the f-sum rule (13) in the long-wavelength limit. In extending the sum rules to higher temperatures it is to be noted that the odd-moment sum rules remain unchanged while Eq. (12) is to be rewritten as

$$S(k)=\frac{|\langle 0|\rho(\vec{k})|1\rangle|^2}{\langle 1|1\rangle}\coth\beta\omega_1(k)/2+\sum_{n\neq 1}\frac{|\langle 0|\rho(\vec{k})|n\rangle|^2}{\langle n|n\rangle}\coth\beta\omega_n(k)/2. \tag{15}$$

By assuming that the single-phonon excitations $|1\rangle$ and the two-particle excitations with frequencies given by Eq. (4) saturate the sum rules, Sridhar and Vasudevan[25] have obtained the single- and two-particle matrix elements

$$|\langle 0|\rho(\vec{k})|1\rangle|^2/\langle 1|1\rangle$$

and

$$|\langle 0|\rho(\vec{k})|2\rangle|^2/\langle 2|2\rangle,$$

respectively, in terms of $V(k)$ and the structure factor $S(k)$. In their approach $S(k)$ itself is given by a linear integral equation involving $V(k)$. Since $V(k)$ is not known precisely, an exact evaluation of the matrix elements is not possible. Furthermore, a realistic potential such as the Lennard-Jones potential that possesses a hard core cannot be used in the integral equation obtained in Ref. 25 since such a potential does not possess a Fourier transform. Hence a solution of $S(k)$ involves the use of a suitably chosen "soft-core" potential.

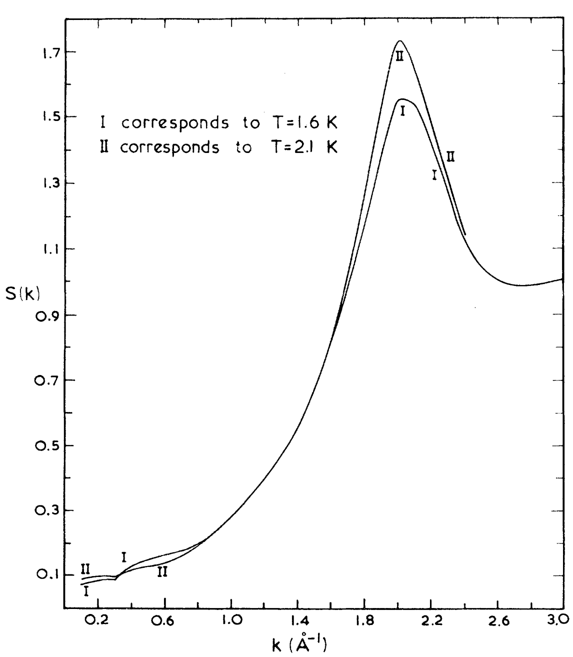

FIG. 1. Structure factor with the use of temperature-dependent excitation spectrum. I corresponds to $T=1.6$ K. II corresponds to $T=2.1$ K.

However, one can extract the required information from the collection of experimental data itself. The experimentally measured single-phonon excitation energies, the single and multiphonon intensities, the single and multiphonon contributions to the f-sum rule, etc., decide the matrix elements. Still, there is some difficulty in writing down the matrix elements for a range of temperatures below T_λ. In this context the fact that the f-sum rule is independent of temperature, type of interaction, and even particle statistics plays a crucial role in obtaining the information. Cowley and Woods[7] have obtained the single and multiphonon contributions to the first moment of $S(\vec{k},\omega)$. Using this experimental information and following the model proposed by Sridhar and Vasudevan, one can assume that the multiphonon contribution to the first

TABLE I. Height of the principal peak of $S(k)$.

	Extrapolated from Ref. 1 for 150.3 kg/m	From the present calculation for 145.3 kg/m^3
$T=1.6$ K	1.445	1.55
$T=2.1$ K	1.505	1.73

moment of $S(\vec{k},\omega)$ is dominated by two-particle excitations. With $\omega_2(k)$ and $\omega_1(k)$ given by Eqs. (4) and (10), respectively, the two-particle and single-particle matrix elements are obtained easily. These matrix elements when used in Eq. (15) yield the structure factor as a function of temperature. In this calculation, the following choice of ω_2 is made: $a=25$ K and $k_0=2.0$ Å^{-1}.

The principal peak of the structure factor is found to be increasing as the temperature is increased from below T_λ towards T_λ (see Fig. 1). The calculated increase in the peak height of 2.1 K as compared to its height at 1.6 K is about 1.5%, while on the basis of experimental information[1] it is expected to be around 4.5%.[26] Furthermore, in this approach there is no appreciable change in the width of the peak.

This defect has been rectified by taking into account the proper temperature dependence of the excitation energies that precisely account for the quasiparticle interactions at nonzero temperatures. Experimental information concerning the temperature dependence of one-phonon excitation energies is readily available for $T=1.1$, 1.6, and 2.1 K.[7] However, one can deduce $\omega_2(k)$ at only one temperature $T=1.1$ K. Thus one is forced to take this value for ω_2 even for the other temperatures.

The incorporation of the proper temperature dependence of the excitation energies brings about a considerable change in the temperature dependence of $S(k)$. The increase in the peak height at 2.1 K as compared to its height at 1.6 K is about 11% as shown in Table I. Although there is a broadening of the peak, quantitative comparison with the experiment cannot be made as this information is not given in Ref. 1.

In addition to the above-mentioned moments one may also consider the inverse-moment sum rule:

$$-\frac{\chi(k)}{2m\mathscr{S}^2}=\int_0^\infty \frac{d\omega}{\omega} S(\vec{k},\omega)=\frac{1}{\omega_1(k)}\frac{|\langle 0|\rho(\vec{k})|1\rangle|^2}{\langle 1|1\rangle}+\sum_{n\neq 1}\frac{1}{\omega_n(k)}\frac{|\langle 0|\rho(\vec{k})|n\rangle|^2}{\langle n|n\rangle}, \tag{16}$$

where $\chi(k)$ is the density-density fluctuation function and $\mathscr{S}$ is the velocity of sound. In the long-wavelength limit

$$\lim_{k\to 0}\chi(k)=-1\,. \tag{17}$$

Following the prescription previously used one can compute the velocity of sound by the use of Eqs. (16) and (17). This is done by suitably rewriting Eq. (16), whose form remains the same even for nonzero temperatures. The velocity of sound and the density correlation function are calculated, in-

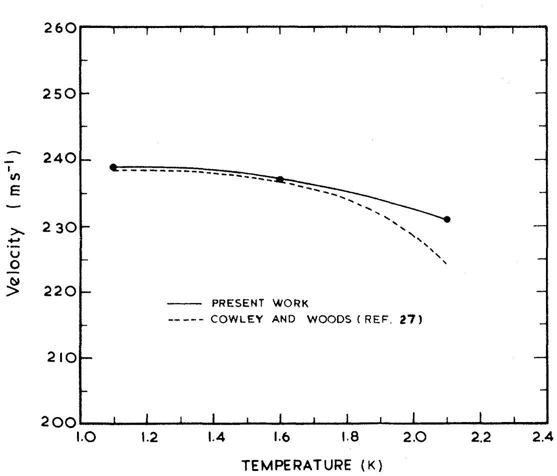

FIG. 2. Velocity of sound vs temperature.

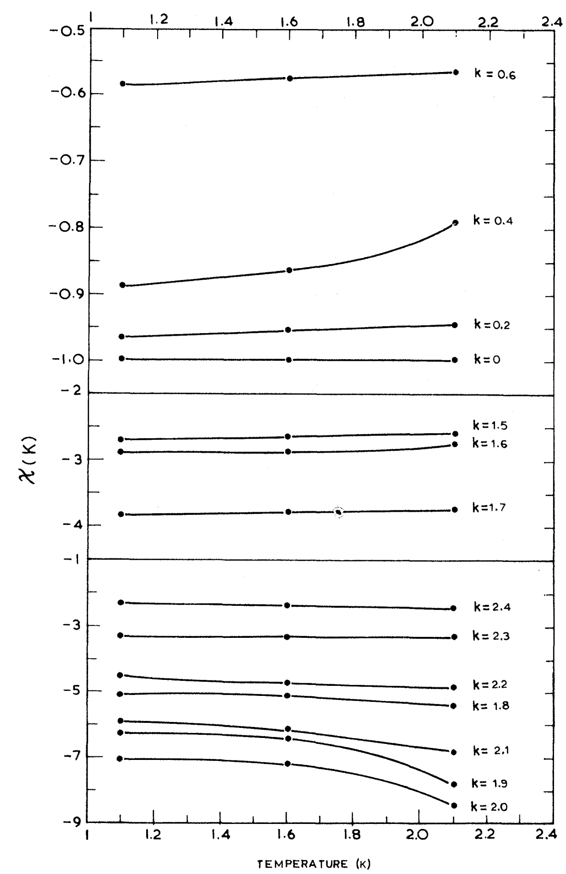

FIG. 3. Density-density correlation: $\chi(k)$ vs T.

serting the proper temperature dependence of the excitation energies; the results are shown in Figs. 2 and 3, respectively.

The velocity of sound is in qualitative agreement with experimental observation—especially ultrasonic measurements (as quoted by Woods and Cowley[27]). The quantitative agreement has to be improved. Regarding the correlation function $\chi(k)$ it is a constant for zero-momentum transfers. For nonzero-momentum transfers $\chi(k)$ indicates that the density-density correlations for momentum transfers of the order of 2 $\mathrm{\AA}^{-1}$ (corresponding to roton excitations) decreases as the temperature is increased from $T=0$ to T_λ.

The suggestion that the loss in spatial order may be due to Bose-Einstein condensation[2] has been critically examined by several investigators. While Griffin[4] has questioned the assumption of Hyland *et al.* that the single-particle correlations are negligible at low temperatures, Chester and Reatto[5] argue that the formula of Hyland *et al.* cannot be derived from the Frohlich decomposition of the two-particle density matrix. Fetter[6] has explicitly demonstrated that a weakly interacting Bose gas at low temperatures (which is supposed to exhibit Bose-Einstein condensation rather conspicuously) itself provides a clear counter example to the form of the pair correlation function proposed by Hyland *et al.* Since it is well known that the technique based on the use of reduced-density matrices may not be valid for such a strongly interacting system as that found in liquid helium, the only other available explanation for the decrease in spatial order has been given by De Michelis *et al.*[3]—the thermal excitation of rotons. This description is based on an explicit model of the density matrix for liquid helium corresponding to the Landau picture of noninteracting phonons and rotons. However, in view of the suggested hybridization of these excitations[16] this model alone may not yield the full mechanism for the loss in spatial order.

Since the roton dip is essentially a quantum effect and characteristic of liquid helium, it is natural to expect that the second diffuse branch of excitations, so far observed only in liquid He II, may also contribute to the loss in spatial order below T_λ. Since the phonon branch is present even in normal liquid He (above T_λ) and is observed in some other liquids as well, the peculiar behavior of the spatial order in He II should be a consequence of the special excitations this quantum liquid can sustain. The fact that the roton excitations and two-particle excitations are related is indicated in the behavior of $\chi(k)$. The density-density correlation function, calculated under the assumption that the sum rules are exhausted by single- and two-particle excitations, shows a special behavior at the wave vector $k=2.0\ \mathrm{\AA}^{-1}$ corresponding to the roton dip (see Fig. 3). (The rate of decrease of $\chi(k)$ for fixed k as a function of temperature is more for values of k of the order of roton minimum.) This indicates therefore, the role played by roton excitations in determining the spatial order along with the two-particle excitations.

In the present contribution the single- and two-particle matrix elements [and hence the interparticle potential $V(k)$] are deduced from the experimentally observed contributions of the single and multiphonon excitations to the f-sum rule. The obtained decrease in the height of the principal peak of $S(k)$ as the temperature is lowered from T_λ is found to be larger than the one that might be expected from experiment. (See Table I.) This is mainly due to the fact that the experimental information regarding $\omega_2(k)$ for temperatures other than 1.1 K is not available.[7,27]

On the basis of the rough estimates presented in this contribution, it can be stated that the two-particle excitations may play a significant role in determining the spatial order in liquid He II. The results presented here are of a preliminary nature since experimental information regarding two-particle excitations is very meager. Thus, to verify the assertions, detailed experimental investigation in this direction is required and this is expected to be fruitful, according to the observations of Blagoveshchenskii *et al.*[15] Theoretically it seems worthwhile to have a microscopic study of the roles played by roton and two-particle excitations in the determination of short-range correlations.

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