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We explain the second branch of excitations in superfluid $^4$He observed by Cowley and Woods, by accounting for two-phonon contributions to the dynamic structure function, $S(k, \omega)$. Our theory gives a good fit with the experimental data in the high energy region for several values of momentum transfer. It is observed that the contribution to $S(k, \omega)$ due to two-phonon excitations is of the order of $k^2$ as against its $k^4$ dependence reported in earlier theories.

Recent inelastic neutron scattering experiments from superfluid helium II by Cowley and Woods [1] show that for certain values of momentum transfer $k$ the spectrum of the dynamic structure factor, $S(k, \omega)$, has, in addition to the sharp one phonon peak, an extra peak observed around an energy transfer $\omega \approx 25$ K. This peak is being attributed to a new mode of excitations of the system, such as the multiphonon excitations. Using diagramatic perturbation theory Family [2] has shown that the variation of $S(k, \omega)$ with $\omega$ in the region of high-energy transfer (deep inelastic region) is independent of the particle statistics and it depends only on the interparticle potentials. In a subsequent paper Wong [3] has tried to explain the high-energy tail of the spectrum of $S(k, \omega)$ at temperature $T = 1.2$ K for $k = 0.8$ Å$^{-1}$, by assuming that this is mainly contributed by multi-(particle/phonon) excitations. In order to get a fit with the inelastic neutron scattering data at high energies he had to introduce two parameters into his theory: one to account for the approximations used and the other for the multi-excitation contributions beyond two. It has been shown by Wong that for small $k$, the double excitation part of $S(k, \omega)$, which hereafter will be denoted by $X_2(k, \omega)$, varies as $k^4$. For short range potentials of the delta function type, he found that the energy dependence of $X_2(k, \omega)$ for large $\omega$ is given by $X_2(k, \omega) \sim \omega^{-1/2}$, whereas for long range potentials, $X_2(k, \omega) \sim \omega^{-11/2}$. Family [2] had also arrived at similar behaviour both for Fermi and Bose systems.

Although Wong has succeeded in explaining the high energy tail of the spectrum of $S(k, \omega)$, the excitation peak around $\omega = 25$ K remains still unexplained. In this paper we have tried to explain this peak by estimating the contribution due to two-phonon excitations. We start with the equation of motion for the density fluctuation operator $\rho_k(t)$ [4], which is written as,

$$\rho_k^+(t) = \sum_{i=1}^{N} \left( \frac{k \cdot p_i}{m} - \frac{k^2}{2m} \right) e^{i k \cdot r_i(t)} + \sum_q \frac{k \cdot q}{m} U(q) \rho_{-k-q}(t)$$

(1)

$$= \sum_{i=1}^{N} e^{-i k \cdot r_i(t)}$$

where

$$\rho_k(t) = \sum_{i=1}^{N} e^{-i k \cdot r_i(t)}$$

In eq. (1) $U(q)$ denotes the Fourier transform of the interparticle potential, and $m$ the mass of the helium atoms. We choose the potential acting between any pairs of atoms in the liquid $^4$He to be of the short range type [5].
where $b$ is the scattering length. The dynamic structure factor $S(k, \omega)$, is defined as,

$$S(k, \omega) = \sum_n \langle \rho_k^n \rangle_0^2 \delta(\omega - \omega_n),$$

(3)

where the indices 0 and $n$ in (3) refer to ground and excited states of the many particle system respectively and $\omega_n = (E_n - E_0)$ is the excitation energy. Since for a fixed $k$ the one phonon excitation spectrum persists up to some value of $\omega$, beyond which the two phonon excitation contribution is likely to start, we assume that the peak in the high energy region of $S(k, \omega)$ is dominantly contributed by the two-phonon (pair) excitations. In eq. (3) this amounts to confining ourselves to values of $n = 2$. Since the first term in eq. (1) is proportional to a single density fluctuation operator, it does not contribute to the matrix element $\langle p_k \rangle_0^2$ when averaged over the states $|0\rangle$ and $|2\rangle$, whereas the second term contributes. Separating out the time dependence from $p_k$ and taking its matrix element over the states $|0\rangle$ and $|2\rangle$, we find,

$$\langle \rho_k^2 \rangle_{20} = \langle 2 | \rho_k^2 | 0 \rangle = \sum_q \frac{1}{\omega_{20}^2} \frac{k \cdot q}{m} U(q) \langle \rho_{-k - q\rho_q} \rangle_{20}.$$  

(4)

Using the inverse Fourier transform for $V(r)$ and the expression for $\rho_k$, eq. (4) can be written after interchanging $i$ and $j$ as,

$$\langle \rho_k^2 \rangle_{20} = -\frac{1}{2} \langle 2 | \frac{1}{m \omega_{20}^2} \sum_{i \neq j} i k \cdot \nabla_{ij} \delta(r_{ij}) [e^{i k \cdot r_i} - e^{i k \cdot r_j}] | 0 \rangle.$$  

(5)

Now expanding $\exp [ik \cdot r_i] \approx 1 + ik \cdot r_i$, and similarly $\exp [ik \cdot r_j]$, which is equivalent to the dipole approximation, (5) can be simplified to give

$$\langle \rho_k^2 \rangle_{20} = \frac{1}{2} \langle 2 | \frac{1}{m \omega_{20}^2} \sum_{i \neq j} (k \cdot r_i)^2 | r_{ij} | u(r_{ij}) | 0 \rangle.$$  

(6)

From (6) one obtains the expression for $X_2(k, \omega)$, which is the same as the one obtained by Wong [3]. Obviously, this gives rise to the $k^4$ dependence of $X_2(k, \omega)$ since it involves $(\langle \rho_k^2 \rangle_{20})^2$. However, we shall try to evaluate the matrix element $\langle \rho_{-k - q\rho_q} \rangle_{20}$ appearing in eq. (4), by writing the state $|2\rangle$ as follows:

$$|2\rangle = \rho_{p_1} \rho_{p_2} | 0 \rangle,$$

(7)

where

$$\rho_p = \sum_{p_3} C_{p_3}^+ C_{p_3 + p}.$$  

(8)

In this $C_{p}^+ (C_{p})$ denotes the creation (annihilation) operator for bosons (corresponding to a helium atom). Using (8) in eq. (7) we now evaluate the matrix element $\langle \rho_{-k - q\rho_q} \rangle_{20}$ by applying Wick’s theorem to the product of boson operators. In this evaluation, retaining the matrix element corresponding to the direct contributions, we find,

$$X_2(k, \omega) = \sum_q \sum_{p_1} \sum_{p_2} \frac{1}{m^2 \omega^4} \frac{k^2}{m} U(q)(k \cdot q)^2 n_{p_1} (1 + n_{p_1 + q + k}) n_{p_2} (1 + n_{p_2 + q})$$

$$\times \delta \left[ \omega \left( q^2 + \frac{k^2}{2m} + \frac{k \cdot q}{m} + \frac{k \cdot p_1}{m} + \frac{q \cdot p_1}{m} + \frac{q \cdot p_2}{m} \right) \right].$$  

(9)
where \( n_p \) denote the distribution functions for the bosons. To make the calculation simpler we replace the terms within the argument of the delta function by their respective averages. As a result, the terms of the type \( q \cdot p \) average to zero. Now, identifying the terms within the brackets \( \sum n_{p_1} n_{p_2} (1 + n_{p_1} + k) n_{p_2} (1 + n_{p_2} + q) \) with \( n^2 S(q) S(q + k) \) where \( S(q) \) denote the static structure function of the system and \( n \) is the average particle density in the system, we write eq. (9) as,

\[
X_2(k, \omega) = \frac{n^2}{2} \sum_q U^2(q) (k \cdot q)^2 S(q) S(q + k) \delta \left( \omega - \frac{k^2}{2m} - \frac{q^2}{m} \right).
\]

(10)

The identification we have made above is done in analogy with the Fermi system. The very choice of writing \(|2>\) in the way shown in eq. (7) can only be justified by ignoring correlations between the particles. We take care of the fluid correlations by replacing the unperturbed static structure functions appearing in eq. (10) with the ones measured experimentally. From eq. (10), it is apparent that the variation of \( X_2(k, \omega) \) with \( k \) is actually of order of \( k^2 \).

For small \( k \), we make a Taylor series expansion of \( S(q + k) \) and deep terms up to order \( k^2 \). It is known that for large \( q \), that is for \( q > q_0 \), a critical value, \( S(q) \) goes to unity. If one is in this large \( q \) region, from the argument of the delta function in eq. (10) one immediately sees that this would automatically restrict one to large \( \omega \) values given by \( \omega = \sqrt{q^2/m + k^2/m^2} \). Therefore, in the region of large \( \omega \), we have the expression for \( X_2(k, \omega) \)

\[
X_2(k, \omega) \approx \frac{n^2}{m^2 \omega^4} \sum_q U^2(q) (k \cdot q)^2 S(q) S(q + k) \delta \left( \omega - \frac{k^2}{2m} - \frac{q^2}{m} \right).
\]

(11)

where \( U = 4 \pi nb/m \). The region of low energy transfer would be determined by values of \( q \) less than \( q_0 \). In this region we choose the form for \( S(q) \) as given by [1]

\[
S(q) = (2mc)^{-1} (q - zq^3),
\]

(12)

where \( c = 237.5 \) m/s, is the isothermal sound velocity in liquid \(^4\)He in the superfluid phase, and the parameters \( z = (1.5 \pm 0.2) \) \( \AA^2 \). For small \( \omega \) the expression for \( X_2(k, \omega) \) is given by,

\[
X_2(k, \omega) = \frac{(n^2/4\pi^2)}{U^2 m^{1/2}} F(k, \omega),
\]

(13)

where

\[
F(k, \omega) = \frac{mk^2}{12c^2} \left[ z^2 \omega^{1/2} - \frac{2z}{m} \omega^{-1/2} + \frac{1}{m^2} \omega^{3/2} \right] + \frac{k^4}{8c^2} \left[ \frac{z^2}{m^2} \omega^{-1/2} + \frac{6z}{m^3} \omega^{3/2} - \frac{7}{m^2} \omega^{5/2} \right].
\]

(14)

From eq. (11) we notice that in the high energy region the dominant contribution from the multi-excitations in liquid \(^4\)He is of order \( k^2 \omega^{-5/2} \) instead of \( k^4 \omega^{-7/2} \) as obtained by Family and Wong both. The frequency dependence of \( X_2(k, \omega) \) is supposed to be different for different forms of potentials. Extending our calculation to charged quantum systems, we notice that for large \( \omega \) our \( X_2(k, \omega) \) varies as \( \omega^{-9/2} \) instead of \( \omega^{-11/2} \), a behaviour which has been reported by Family [2] and Wong [3]. We have made a calculation of the intensity of the scattered neutrons, for \( k = 0.6 \) \( \AA^{-1} \) and \( k = 0.8 \) \( \AA^{-1} \) in the high energy region using expression (11), starting from \( \omega = 70 \) K and going to lower values. For such momenta the intensity in the relatively low energy region is calculated from eq. (13). On extrapolating the curves from both sides we notice that they meet at a point where \( \omega \approx 22.5 \) K for \( k = 0.6 \) \( \AA^{-1} \) and \( \omega \approx 22.1 \) K for \( k = 0.8 \) \( \AA^{-1} \). Both these numbers are very close to values of \( \omega \) at the positions of the second peaks that have been observed in the neutron scattering experiments [1] (which roughly come out to be 20.07 K and 24.5 K respectively). Our results are shown along with the experimental data, in figs. 1 and 2. From the graphs we find that our theoretical results agree very well with the experiment both in the high and low energy transfer region. It is interesting to notice that we have not used any adjustable parameters in our theory like those introduced by Wong [3] in order to obtain a fit with the experiment.

Since our calculations indicated that the contribution from the two-phonon excitations varies as \( k^2 \) it is obvious to think that the one phonon excitation does not exhaust the f-sum rule to order \( k^2 \). It will be interesting
to study the implication of such a dependence of the two-phonon excitations on the various properties of the system.

AN EXPLANATION OF THE STRUCTURE IN THE DYNAMIC STRUCTURE FACTOR
OF AN ELECTRON LIQUID

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We explain the recently observed interesting structure in the dynamic structure factor \( S(k, \omega) \) of various metallic systems by considering processes that involve single-particle and multi-particle excitations.

Recent inelastic X-ray and electron scattering experiments from metals, particularly those of Platzman and Eigenberger \([1–3]\), have revealed an interesting double-peak structure in the dynamic structure factor, \( S(k, \omega) \), for momentum transfers lying within \( k_c \leq k \leq 2k_F \), where \( k_c \) is the critical wave vector up to which plasmons exist as well-defined excitations of the system and \( k_F \) is the Fermi vector. The above feature in \( S(k, \omega) \) has been observed in a wide variety of different systems including metallic Al, Be, Li and graphite, etc. Since this feature in all these systems with very different band structures, looks similar, it is suggested that this is perhaps due to some phenomenon associated with the dynamical behaviour of the interacting electron gas.

To explain the structure in \( S(k, \omega) \) it is argued that the spectrum in the above momentum transfer region is composed of a spectrum due to plasmon excitations plus one due to electron–hole pair excitations. It therefore amounts to first proving that the plasmon modes continue to exist for values \( k > k_c \). Without caring for the existence of the plasmon modes to the region beyond \( k_c \), there have been attempts \([4]\) to calculate \( S(k, \omega) \) using the dielectric function of the random phase approximation (RPA) and that of Vashisht and Singwi (VS), which goes beyond the RPA. These calculations do not indicate any structure in \( S(k, \omega) \). Although in a later work by Mukhopadhyay et al. \([5]\) which makes use of a modified form of the VS dielectric function by incorporating lifetime effects into the theory, there seems to have appeared some structure in \( S(k, \omega) \), it has been shown by Rao et al. \([6]\) that a rigorous treatment of lifetime effects in the type of calculations done by Mukhopadhyay et al., does wipe out the structure in \( S(k, \omega) \). It is worth mentioning a more recent calculation of \( S(k, \omega) \) by de Raedt and de Raedt \([7]\) that is based on Mori’s memory function formalism. There are certain basic objections against this theory \([6]\). Besides, since this theory deals with several adjustable parameters to explain the structure, one cannot see what effects are actually responsible for giving rise to the structure. Very recently Barnea \([8]\) has been able to reproduce the structure by considering a process in which a plasmon decays into an electron–hole pair plus a plasmon in addition to the usual RPA single-pair decay. This theory makes use of an ansatz to evaluate the matrix element associated with the plasmon decay into an electron–hole pair and a plasmon. The basic question that plasmons exist beyond \( k_c \) remains unanswered here also. Besidés, this theory has the unphysical feature of having negative values of \( S(k, \omega) \) for higher frequencies. In a recent work it has been shown by Pal et al. \([9]\) that the plasmon spectrum actually extends to regions beyond \( k_c \). In this work the authors have extracted the plasmon contribution to the electron–hole pair from beyond the random phase approximation due to the exchange of virtual plasmons and added this to the plasmon contribution in the RPA. With this they have reproduced the experi-
mentally observed frequency shift to classical plasma frequency at $K = 0$, by redefining $k_c$. This has resulted in arriving at a new value for $k_c$, which is much larger than the $k_c$ given by RPA. In a later work by Tripathy and Pal [10] it has been shown by means of a calculation of the dynamic structure factor corresponding to the plasmons and then adding to this the structure factor corresponding to particle–hole excitations, that there is no structure in $S(k, \omega)$ for wave vectors close to the experimental values. Rather, it is seen from this calculation that there is the indication of some structure of the plasmon and quasiparticle excitations but due to something else. In the present work we have tried to explain the structure by visualising that this arises because of the combination of one-pair and multipair excitation spectrum. In particular, we have calculated the two-pair excitation contribution to the dynamic structure factor and added this to that of the one-pair excitations. With this we find that the structure in $S(k, \omega)$ for several systems like Al, Li and Be is more or less reproduced.

To calculate the two-pair excitation contribution to $S(k, \omega)$, we start with the equation of motion for the density fluctuation operator, $p^+(t)$ [11], which is written as

$$\rho_k(t) = -\sum_{i=1}^{N} \left( \frac{k \cdot p_i}{m} + \frac{k^2}{2m} \right) \exp[ik \cdot r_i(t)]$$

$$+ \sum_q (k \cdot q/m) U(q) \rho_{-k-q}(t) \rho_q(t), \quad (1)$$

where $U(q) = 4\pi e^2|q|^2$, the Fourier transform of the interparticle potential. The dynamic structure factor of the system of an electron gas is defined as

$$S(k, \omega) = \sum_n |\langle \rho_k^+ \rangle_n \rangle^2 \delta(\omega - \omega_n), \quad (2)$$

where $|0\rangle$ and $|n\rangle$ refer to the ground and excited states of the system and $\omega_n$ corresponds to the excitation energy. The excited states $|n\rangle$ constitute of states of one, two and higher pairs of excitations of the system. Here we are interested in the contributions to $S(k, \omega)$ coming from two-pair excitations. As evident from (1) the first term on the right-hand side does not contribute to two-pair excitations whereas the second term does. From this equation, after separating out the time dependence using the property of invariance under time translations, we obtain following (1),

$$\langle \rho_k^+ \rangle_{20} \langle 2| \rho_k^+ |0\rangle$$

$$= - \sum_q \omega_2^{-2} (k \cdot q/m) U(q) \langle 2| \rho_{-k-q} \rho_q |0\rangle. \quad (3)$$

The above matrix element is evaluated using the method of quantum field theory according to which one writes

$$\rho_k = \sum_{p, \sigma} C^\dagger_{p, \sigma} C_{p+k, \sigma}, \quad (4)$$

where $C^\dagger_{p, \sigma}$ ($C_{p, \sigma}$) denotes the creation (annihilation) operator for the electrons $\sigma$ being referred to the spin index. It can be shown that the contribution to $\langle \rho_k^+ \rangle_{20}$ is evaluated by writing the state $|2\rangle$ as

$$|2\rangle = \rho_{p_1^+} \rho_{p_2^+} \rho_{p_1^-} \rho_{p_2^-} |0\rangle. \quad (5)$$

Using (4) and applying Wick's theorem to the product of Fermi operators we write the two-pair contribution to $S(k, \omega)$ as,

$$S_{2}(k, \omega) = \sum_{q, p_1, p_2} (4/\omega^{4}) U^2(q)(k \cdot q/m)^2 \times n_{p_1}(1 - n_{p_1^+ + k_1} n_{p_2}(1 - n_{p_2^+ + q})$$

$$\times \delta \left( \omega - \left[ \frac{q^2}{m} + \frac{k^2}{2m} + \frac{k \cdot q}{m} + \frac{k \cdot p_1}{m} + \frac{p_1 \cdot q}{m} + \frac{q \cdot p_2}{m} \right] \right) \quad (6)$$

where $n_p$ denotes the distribution function for the fermions, the suffix 2 in $S$ refers to the two-pair excitations. Eq. (6) denotes the matrix element corresponding to the direct processes. In order to avoid complications involved in dealing with angular integrations we approximate the terms within the argument of the delta function by their angular average values. As pointed out by Woods and Cowley [12] the terms of the type $q \cdot p$ give no contribution to the peak position but are responsible for width about $q \cdot p/m$. To compensate for using such an approximation we introduce a parameter $\alpha$ into our theory replacing the factor $k^2/2m$ by $\alpha k^2/2m$. The parameter

\[253\]
\( \alpha \) has been used to take care of the \( k \) dependence of the terms that are averaged out to zero. The value of this parameter is fixed from a best fit with the experimental data. With the above simplification (6) reduces to the form,

\[
S_2(k, \omega) = \left( \frac{n^2}{m^2} \omega^4 \right) \sum_q U^2(q)(k \cdot q)^2 S(q)S(q + k) \times \delta(\omega - q^2/m - \omega k^2/2m), \tag{7}
\]

where \( S(q) \) denotes the static structure factor of the system.

In the absence of any analytical form of \( S(q) \) either in RPA or beyond, we shall be using its Hartree-Fock value \([13]\) for evaluating the integral given in (7). An estimation of \( S_2(k, \omega) \) as a function of \( \omega \) is done numerically for systems like Li, Be, Al metals and graphite, corresponding to the values of wave vectors for which experiments have been performed. Using these values for \( S_2(k, \omega) \) the total \( S(k, \omega) \) is determined by adding this to \( S_1(k, \omega) \) the one-pair excitations part. For \( S_1(k, \omega) \) we have also chosen its Hartree-Fock value. We have plotted \( S(k, \omega) \) against \( \omega \) for different values of \( K \) and compared our results with those of the experiments for Al, Be, Li and graphite. These are shown in fig. 1. The value of \( \alpha \) needed to obtain a good fit with the experimental data is found to be 1.4 for Al, 1.5 for Be and graphite, 1.8 for Li. It may be mentioned here that with \( \alpha = 1 \) which is equivalent to replacing the terms in the argument of the delta function in (6) by their angular averages we are also able to observe the double peak structure in the total \( S(k, \omega) \). However the height of the right-hand peak happens to be larger than that of the left-hand peak. This is in contrast with experimental observation. Now adjusting the value of \( \alpha \) to a value greater than unity the peak heights are made similar to the experimental ones. In order to check whether with the use of a better form of \( S(q) \) our calculated structure in \( S(k, \omega) \) remains or not, we have performed a calculation of \( S_2(k, \omega) \) for Be using the \( S(q) \) of the random phase approximation (RPA). Since the analytic form for \( S(q) \) in the RPA is not available we have first tried to obtain a fitted form of \( S^{\text{RPA}}(q) \) for Be. Using this fitting formula, we have calculated \( S_2(k, \omega) \) for \( k = 1.46k_F \) and \( 1.76k_F \) for the case of Be. These are added to the corresponding \( S_1(k, \omega) \) of the RPA to
Fig. 1. Plots of $S(k, \omega)$ versus $\omega/\epsilon_F$ for Be, Al, Li and graphite for fixed values of $k$. The peak heights of all curves have been normalised to coincide with the experimental peaks. The solid curves are the ones determined from the present theory with HF input for $S(q)$ and $S_1(k, \omega)$. The dashed curves represent those calculated from the present theory with $S(q)^{\text{RPA}}$ and $S_1^{\text{RPA}}(k, \omega)$. The dashed curves with intermediate dots represent the experimental ones.
obtain the total $S(k, \omega)$. It is to be noted that for both $k = 1.46k_F$ and $1.76k_F$ we also observe the two-peak structure in $S(k, \omega)$ as before. However $\alpha$ now takes a slightly different value, i.e. 1.3 against 1.5 chosen earlier. These are shown in the graph for Be.

From the graphs we notice that our calculated $S(k, \omega)$ more or less agree with the ones observed from experiments for almost all the systems. We find that the peak positions of our $S(k, \omega)$ do not match with those obtained from experiments. Such deficiency in our theory may be due to the fact that we have taken $S_{HP}(k)/S_{RPA}(k)$ as the input to calculate $S(k, \omega)$. We think that the first peak in $S(k, \omega)$ corresponds to the energy for the one-pair excitation, whereas the second peak mostly accounts for the two-pair excitation processes. The energy difference between the two peak positions in our theory almost agrees with that given by the experiments. The tailing feature of $S(k, \omega)$ in our theory results from two-pair excitations. It is to be noted that for very low and very high values of momentum transfer $k$ we do not observe structure in $S(k, \omega)$ with $\alpha = 1$ (no parameter case) by adding the contribution of $S_{2}(k, \omega)$ to $S(k, \omega)$. In such cases adjustment of the values of $\alpha$ does not arise. In the absence of any means of finding $\alpha$, if we choose the same $\alpha$ for both high and low $k$ values there appears no structure in $S(k, \omega)$. We have made a calculation of $S(k, \omega)$ at $k/k_F = 2.1$ for the case of Be, which shows no structure with $\alpha = 1.5$. This is also shown in the graph. In the case of Be it is seen that there is an additional structure in $S(k, \omega)$ at the low-energy side, that is at $\omega = 1.3 e_F$, for $k = 1.76k_F$. As we find from the graph our theory is not able to reproduce the new structure. The reason for this is not clear to us at present.

We conclude by saying that the observed structure in $S(k, \omega)$ can be explained assuming it to be due to the superposition of one-pair and two-pair excitations. The fact that it is due to the addition of a plasmon to a quasiparticle pair spectrum is almost ruled out in our theory. This is because it is hard to believe that a plasmon does exist for wave vectors close to $2k_F$, unless one shows that the new critical wave vector $k_c$ is much larger than its RPA counterpart.

References

In a recent paper [1] (hereafter to be referred to as I) we have tried to explain the second branch of elementary excitations observed by Cowley and Woods in neutron scattering experiments [2] from superfluid \(^4\)He by evaluating the two-phonon excitation contribution to the total dynamic structure factor \(S(k, \omega)\) of the system. From our calculation of the two-phonon excitation spectrum denoted by \(S_{II}(k, \omega)\) it is observed that this exhibits a peak at a value of \(\omega\) which is very close to the position of the second peak observed in \(S(k, \omega)\), when plotted as a function of \(\omega\), for fixed values of momentum transfer \(k\). Because of this we conclude that the second-branch excitations observed by Cowley and Woods [2] are due to two-phonon excitations in the system. Looking at the expression for \(S_{II}(k, \omega)\) derived by us we find that in the region of high energy transfer the dominant contribution to \(S_{II}(k, \omega)\) in the case of liquid \(^4\)He goes as \(k^2/\omega^{5/2}\) instead of the behaviour \(k^4/\omega^{7/2}\) obtained by Family [3] and Wong [4]. Such a form of variation with \(\omega\) is only possible for a potential between a pair of helium atoms of the form

\[
V(r) = (4\pi b/m)\delta(r),
\]

where \(b\) is the scattering length and \(m\) is the mass of the helium atom. For a charged quantum liquid, the high energy behaviour of \(S_{II}(k, \omega)\) has been shown by us to be given by \(k^2/\omega^{9/2}\) instead of \(k^4/\omega^{11/2}\) derived by the above two workers [3,4]. Such type of high energy behaviour of \(S_{II}(k, \omega)\) has been shown to be a universal property of the system irrespective whether the particles obey Fermi–Dirac or Bose–Einstein statistics. Rather this is purely given by the form of the potential between a pair of atoms in the system.

Since our calculation of \(S_{II}(k, \omega)\) indicates that the contribution from the two-phonon excitations varies as \(k^2\), rather than a \(k^4\) dependence as reported in earlier theories, it is natural to think that the one-phonon excitation alone cannot exhaust the \(s\)-sum rule [2] to order \(k^2\). In the present work we make an estimation of the two-phonon contribution to the \(s\)-sum rule for a set of \(k\)-values. Using the expression for our \(S_{II}(k, \omega)\), we have calculated the static structure factor \(S_I(k)\) of the system as a function of \(k\). The values of \(S_{II}(k)\) so obtained are fitted with a polynomial [5] which is written in powers of \((hk/m)s\), where \(s\) is the isothermal sound velocity. Here we find that the coefficient of the term \(k^2\) is finite instead of zero as given in earlier theories. Some further consequences of the presence of the \(k^2\) term are also discussed in this paper.
To start with, we consider the frequency moments of $S(k, \omega)$, which are defined as

$$\langle \omega \rangle_l = n^{-1} \int_0^\infty \omega^l S(k, \omega) \, d\omega,$$

where the index $l$ refers to the various moments and $n$ denotes the average particle density in the system. For zero temperature the range of $\omega$ integration in (2) is $0 < \omega < \omega_0$. Of the various moments we confine ourselves to $l = 0$ and $1$, in which case we have

$$\langle \omega \rangle_0 = n^{-1} \int_0^\infty \omega S(k, \omega) \, d\omega = \hbar k^2/2m,$$

$$\langle \omega \rangle_1 = n^{-1} \int_0^\infty (\omega^2 S(k, \omega) \, d\omega = \hbar k^2/2m,$$

where $S(k)$ denotes the total structure factor of the system. Eq. (4), which denotes the first moment is known as the f-sum rule. This is a very important relation which follows as a direct consequence of particle conservation in the system. Higher moments are of limited interest to us since it is difficult to determine them experimentally.

We recall the definition of $S(k, \omega)$,

$$S(k, \omega) = \sum_{s'} |(\rho_k^{s'})_0|^2 \delta(\omega - \omega_{s'}) ,$$

where $\rho_k(t)$ is the density fluctuation operator, the indices $0$ and $s'$ refer respectively to the ground and excited states of the many-particle system, which constitutes the liquid $^4$He. In the present case, and $\omega_{s'} = (E_{s'} - E_0)$ is the excitation energy. Since in the region of high energy the two-phonon (pair) excitation contribution to $S(k, \omega)$ is expected to be large, in eq. (5), we confine ourselves to the case $s' = 2$. Using the equation of motion for $\rho_k^{s'}(t)$ and retaining the two-phonon (pair) contribution term which is equivalent to evaluating the matrix element $(\rho_k^{s'})_0^2$, we have arrived at an expression for $S_{11}(k, \omega)$. This was approximated as

$$S_{11}(k, \omega) = \frac{n^2}{m^2 \omega^4} \sum_q V^2(q)(k \cdot q)^2 S(q) S(q + k) \delta(\omega - k^2/2m - q^2/2m) ,$$

where $S_{11}(k, \omega)$ with $k$ is actually of order $k^2$. As shown by us [1], by invoking the dipole approximation in the matrix element for $(\rho_k^{s'})_0$ evaluated by us we arrive at an expression for $S_{11}(k, \omega)$ which is the same as that obtained by Wong [4] where it varies as $k^4$. Because of the $k^4$ variation it was considered that the one-phonon excitation had exhausted the f-sum rule to order $k^2$. However, since we observe a $k^2$ dependence of $S_{11}(k, \omega)$, there would be naturally a contribution to the f-sum rule resulting from the two-phonon (pair) excitations to order $k^2$. Using the form of $S(k)$ for small $k$ and its property for large $k$ we obtain the expression for $S_{11}(k, \omega)$ as

$$S_{11}(k, \omega) = \frac{n^2/4\pi^2}{m^1/2 \nu^2} \times \left[ \frac{k^2}{\omega^{5/2} - (1/4m)k^4/\omega^{7/2}} \right] ,$$

which is valid in the region of large $\omega$. This is identified as the deep inelastic scattering region. In the small-$\omega$ region, we have $S_{11}(k, \omega)$ written as

$$S_{11}(k, \omega) = \frac{n^2/4\pi^2}{m^{1/2} \nu^2} F(k, \omega) ,$$

where $F(k, \omega)$ is some complicated function of $k$ and $\omega$ that is given in ref. [1].

To evaluate $S_{11}(k)$ we make use of eq. (3). In this the lower limit of $\omega$ is determined from the fact that $S_{11}(k, \omega)$ is zero below such a value of $\omega$ for any fixed $k$. We denote this value by $\omega_1$. Thus, as regards the estimation of $S_{11}(k, \omega)$ we break up the integration over $\omega$ as follows:

$$Q \int_0^\infty \omega \, d\omega = \int_0^{\omega_2} \omega \, d\omega + \int_{\omega_2}^\infty \omega \, d\omega ,$$

where $\omega_2$ corresponds to the value of $\omega$ at which the high and low energy curves in our theory meet. This should in principle be given by the two-phonon excitation frequency. We have evaluated $S_{11}(k)$ for values of $k = 0.3, 0.6, 0.8$ and $1.0 \text{Å}^{-1}$, which are shown in table 1. We now try to make a fit of these $S(k)$ values with a polynomial of the form

$$S(k) = S_0(k) + S_{11}(k) = \frac{\hbar k^2}{2m} \left[ 1 + 2z_2(\hbar k/2ms) + z_3(\hbar k/2ms)^2 \right] + z_4(\hbar k/2ms)^3 + ... ,$$

$s$ being the isothermal sound velocity in superfluid $^4$He. The above form of $S(k)$ is written using the expansion

388
Table 1
S_{II}(k) for different k values.

<table>
<thead>
<tr>
<th>k (Å⁻¹)</th>
<th>S_{II} up to k²</th>
<th>S_{II} up to k⁴</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.3</td>
<td>0.00425</td>
<td>0.00429</td>
</tr>
<tr>
<td>0.6</td>
<td>0.01736</td>
<td>0.017572</td>
</tr>
<tr>
<td>0.8</td>
<td>0.0311</td>
<td>0.0314785</td>
</tr>
<tr>
<td>1.0</td>
<td>0.0554</td>
<td>0.048</td>
</tr>
</tbody>
</table>

The formula for the structure function corresponding to the excitation of a single phonon, and that for S_{II}(k) which we have shown to vary as k². Comparing our calculated values for S_{II}(k) with those obtained from (10) we find that the expansion coefficient z₂ associated with the factor \(\frac{1}{2}(\hbar k/m\nu)^2\) has a value \( \approx 0.21 \). This value of z₂ has been seen to remain the same for all the four values of k for which S_{II}(k) has been evaluated. This coefficient z₂ was obtained to be zero in many earlier theories \([3,4,6,7]\), whereas the present theory gives a value \( \approx 0.21 \). The coefficients of the terms \(\frac{1}{2}(\hbar k/m\nu)^3\) and \(\frac{1}{2}(\hbar k/m\nu)^4\) in eq. (10) which have been denoted by z₃ and z₄ respectively have the values \( \approx 3.5 \) and \( \approx 5.8 \), when expressed in dimensionless units. On comparing our estimated value of z₂ \( \approx 0.21 \) with the values of z₃ and z₄ we find that z₂ is actually very small. We have also tried to compare the coefficient of the k⁴ term in S(k) with that of k² for other systems. Following Family and Gould \([8]\) we find that the coefficient of k⁴ divided by the coefficient of the k² term is 2.82 for a charged Bose gas, to be compared with the value \( \approx 27.6 \) obtained by us for superfluid \( ^4\)He, a neutral Bose gas. Therefore from our result it looks that the coefficient of k⁴ is smaller than the coefficient of k² in charged as well as neutral Bose systems. The very estimation of z₂ made above is a direct consequence of the fact that S_{II}(k) varies as k². Since such a variation of S_{II}(k) with k has been missed by several workers one might think it to be an outcome of the approximation that has led us to write S_{II}(k) in the form shown in eq. (6). In order to check this we have further tried to derive S_{II}(k) starting from an altogether different consideration. The new derivation of S_{II}(k) is based on the knowledge of the two-phonon propagator, which is analogous to the expression for the two-plasmon propagator obtained by Deo and Tripathy \([9]\) some time ago. Here we use a definition of S_{II}(k, ω) similar to that of S_{I}(k, ω):

\[ S_{II}(k, \omega) = -\eta^{-1} \text{Im} F_0(k, \omega), \]

where F_0(k, ω) denotes the two-phonon propagator. Following ref. \([9]\) we write

\[ F_0(k, \omega) = n^{-2} \int \frac{dp_1}{(2\pi)^4} d\omega_1 R_2(p_1, \omega_1) \]

\[ \times R_2(p_1 + k, \omega + \omega_1), \]

where

\[ R_2(p_1, \omega_1) = \lim_{\eta \to 0^+} \frac{-\omega(p_1)}{2V(p_1)} \frac{2\omega(p_1)}{[\omega(p_1) - i\eta]^2 - \omega^2}. \]

This is the expression for the one-phonon propagator where \(\omega(p) = sp\), and \(V(p)\) is a constant, \(s\) being the isothermal sound velocity. \(k_c\) represents some critical wave vector up to which the one-phonon exists as a well-defined mode. Evaluating the expression for F_0(k, ω), we determine S_{II}(k, ω). Using this result we obtain S_{II}(k) which is found to have a k² term. The coefficient of k² when estimated gives a value which is of the same order as that obtained by us before.

As we have seen, since S_{II}(k, ω) varies as k² it might contribute to the f-sum rule to order k². To estimate its contribution we use (4) to write

\[ (\hbar k^2/2m)^{-1} \eta^{-1} \int_0^\infty \omega S(k, \omega) d\omega = 1. \]

Following the expression for S_{II}(k, ω) in the high and low energy transfer region we estimate the value of the integral on the left-hand side of eq. (14) for the set of k values used for evaluating z₂. Those are shown in Table 2. From this table we notice that the value of

Table 2
H_{II} for different k values.

<table>
<thead>
<tr>
<th>k (Å⁻¹)</th>
<th>H_{II} up to k²</th>
<th>H_{II} up to k⁴</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.3</td>
<td>0.44548</td>
<td>0.44408</td>
</tr>
<tr>
<td>0.6</td>
<td>0.44918</td>
<td>0.439</td>
</tr>
<tr>
<td>0.8</td>
<td>0.45054</td>
<td>0.43082</td>
</tr>
<tr>
<td>1.0</td>
<td>0.462966</td>
<td>0.405152</td>
</tr>
</tbody>
</table>
the above integral which we have denoted by $H_{j}$ is less than $H_{i}$ for $k < 1 \text{Å}^{-1}$, where $H_{1}$ is obtained by replacing $S_{j}(k, \omega)$ by $S_{i}(k, \omega)$ in (14). This result is in contrast to what has been known from earlier work. Since the value of our $H_{1}$ is found to be decreasing with increasing $k$, this indicates that beyond certain $k$ the contribution to $H_{1}$ would be vanishingly small. This is manifested in the fact that the height of the second peak observed in the neutron scattering experiment almost disappears at larger $k$ values. If this be the case $H_{1}$ would dominate over $H_{1}$ for large $k$ values which would finally approach unity beyond a certain critical value of $k$. This is what is observed for a quantum liquid like the system of an electron gas at $T = 0$ [10]. Because of this we do not see any reason why the system of superfluid $^{4}$He would behave differently from an electron liquid at $T = 0$.

To summarise, we have shown in this paper that the one-phonon excitations in the superfluid $^{4}$He system do not alone exhaust the $f$-sum rule to order $k^{2}$, $S_{1}(k, \omega)$ evaluated by us is also seen to contribute to the $f$-sum rule. Our estimated value for the coefficient $z_{2}$ is found to be finite instead of being zero as known from earlier theories.

One of the authors (M. Bhuyan) thanks the Education and Youth Services Department of Government of Orissa for granting her study leave and financial support.

References

It is found that the lifetime of positrons in the superconducting Aluminium is \( \sim 10^{-8} \) s compared to its value \( \sim 10^{-7} \) s obtained for the normal state. The annihilation rate is seen to increase slowly as the temperature of the system varies, from \( T = 0 \) to \( T = T_c \). The reason for having a very low annihilation rate in the superconducting state is discussed and the experiment for measuring this is suggested.

The positron annihilation rate is known to be very sensitive to the electron wave function at the positron site. Therefore, phenomena like phase transition which is associated with changes in the electronic wave function of the system, is supposed to be studied by means of the positron annihilation techniques. Although there has been a large number of data on the life time of positrons in several materials undergoing varieties of phase transitions, a theoretical understanding of any of these has not been clearly achieved so far. In the present work we have tried to estimate the positron annihilation rate in superconductors where the phase transition has been known to be of second order and also is fairly well understood theoretically. On physical grounds it was long back speculated by Dresden [1] that the mean life time of positron in a superconductor is to be larger than that of the same system in its normal state. Following this suggestion a measurement of the life time of positrons in Pb was made by Stump and Talley [2] which actually indicated larger value of the mean lifetime in the superconducting state than in the normal state. However, the latter measurements of the mean lifetime in superconducting Pb by Shafroth and Marcus [3] by Green and Madensky [4] did not report any noticeable change in the life time from the value observed in the metallic Pb. With a view to have a clear understanding of the true picture we have evaluated the mean life time of positron in superconducting Al and compared our result with its normal state value. We start with the expression for the annihilation rate \( R \) which is written as [5],

\[
R = n \lambda ,
\]

where \( \lambda = \pi r_e^2 c \), and \( n \) is the electron density at the positron site. The determination of \( n \) is done by using the relation:

\[
n = (-i)^2 \int \frac{dk}{2\pi} \frac{G_{\text{ep}}(\mathbf{x}_t, \mathbf{x}_t', \mathbf{x}_t', \mathbf{x}_t')}{t' - t + 0} ,
\]

in which \( G_{\text{ep}} \) denotes the electron-positron Green's function. We make use of the high density expansion for \( G_{\text{ep}} \) which gives rise to the so-called ladder diagram [6] in the case of normal metals. The first order ladder diagram is in fact known to give the most dominant contribution to the positron annihilation rate for normal metals. Here we have to evaluate \( n \) for the situation where the electron belongs to the superconducting state. Confining ourselves to the ladder diagram the expression for \( n \) becomes

\[
n = -\frac{2i}{(2\pi)^4} \int dk \frac{1}{\omega - E_k + i\eta} \chi_0(k,\omega) ,
\]

where \( \chi_0(k,\omega) \) denotes the propagator for the electron-hole pair, and \( U(k,\omega) \) the effective interaction between the electron and the positron. For the superconducting state one obtains

\[
\chi_0(k,\omega) = \sum_p \left( \frac{U_p P_+ + U_{-p} P_-}{E_p - E_{-p}} \right)^2 \frac{1}{\omega + E_p + E_{-p} - i\eta} \]

(6)

\[
\chi_0(k,\omega) = \frac{1}{\omega - E_p - E_{-p} + i\eta} \]

(7)
where the symbols used in Eq. (4) have their usual meanings. The above expression is valid in the neighbourhood of extremely low temperatures \( T < T_c \). In Eq. (3) replacing \( U(k,\omega) \) by its static limit, we arrive at

\[
n = (2\pi)^3 \int d\mathbf{k} U(k,0) \frac{2}{\mathcal{P}} \left[ \sum_{\mathcal{P}} (1 - \frac{\xi_{\mathcal{P}}^+ + \xi_{\mathcal{P}}^-}{E_{\mathcal{P}}^+ + E_{\mathcal{P}}^-}) \right] \frac{1}{\epsilon_{k} + E_{\mathcal{P}}^+ + E_{\mathcal{P}}^-}
\]

where \( \Delta \) is the superconducting gap parameter.

To evaluate the integrals in Eq. (5) for the superconducting state we use the fact that \( (2\pi)^3 \int dp = N(0) \int d\varepsilon \), where \( N(0) \) denotes the density of states at the Fermi level. After evaluating, it is found that the annihilation rate \( J \) in the superconducting Al is \( 0.20 \times 10^9 \text{s}^{-1} \), which is reduced by a factor of 10^4 with respect to the value in the metallic Al \( [R(\text{Al}) = 10^7 \text{s}^{-1}] \). It is further noticed that \( R \) increases by raising the temperature of the system from \( T = 0 \) to \( T = T_c \) \( (= 1.2 \text{ K}) \):

\[
R(0 \text{ K}) = 0.19 \times 10^5 \text{s}^{-1},
\]
\[
R(1 \text{ K}) = 0.26 \times 10^5 \text{s}^{-1}.
\]

In the limit of \( \Delta = 0 \), we obtain the value \( R \sim 10^5 \text{s}^{-1} \) at \( T = 0 \). This is the result for the normal state of Al. In the normal state if the temperature is allowed to increase, the value of \( R \) shows a decreasing result.

The value of the annihilation rate in the superconducting state of metal is found to be reduced by a factor of \( 10^4 \) than its value in the normal state. Physically, the occurrence of such a small value for the annihilation rate in a superconductor is realizable. Because, when the system becomes superconducting one finds that the sharp fermi surface of the normal state at \( T = 0 \) is smeared out over a thickness of 2 in the energy space. The fraction of the total number of electrons, which are close to the Fermi surface only participates in the pairing process and hence contributes to the phenomenon of superconductivity. Since this fraction is \( \approx 10^{-6} \), a great reduction in \( R \) is understandable. Such an extremely low annihilation rate should result in giving rise to a very large lifetime in the superconducting state. This being the case, why is it that one is not able to see any difference in the lifetime of a positron in the superconducting state from its value in the normal state? The reason for this is that one is not able to isolate the contribution to the annihilation rate arising solely from the superconducting electrons. To overcome this difficulty we suggest a two-parameter lifetime measurement, which may enable one to measure the momentum dependent annihilation rate \( R(p) \). In this experiment, if one selects a small window along the momentum axis around \( k \), one may be able to measure the annihilation rate coming from the superconducting electrons only.

REFERENCES: