PHYSICAL REVIEW A VOLUME 1, NUMBER 6 JUNE 1970

High-Energy Neutron Scattering from Liquid He^4. II. Interference and Temperature Effects

V. F. Sears
Atomic Energy of Canada Ltd., Chalk River Nuclear Laboratories, Chalk River, Ontario, Canada
(Received 3 November 1969)

The scattering of high-energy neutrons from liquid He^4 was discussed in an earlier article on the basis of the Gram-Charlier series expansion of the incoherent scattering function. In the present paper, interference effects ignored previously are taken into account approximately and are shown to give rise to an oscillation in the half-width of the scattering function as a function of the momentum transfer. The temperature dependence of the scattering is investigated by deriving approximate expressions for the parameters in the Gram-Charlier expansion in terms of their values at $T=0$, together with the observed thermodynamic properties of liquid He^4. Reasonable agreement with the neutron-scattering data of Cowley and Woods is obtained.

I. INTRODUCTION

In a recent article,[1] the scattering of high-energy neutrons from liquid He^4 was discussed in terms of the Gram-Charlier series expansion of the scattering function. The coefficients in this expansion were denoted by $\epsilon_n(\kappa)$, with κ being the momentum transfer, and it was shown that if $\epsilon_n(\kappa)$ is replaced by $\epsilon_n(\infty)$, the series can be summed exactly to yield the impulse approximation (IA). The IA is, therefore, asymptotically correct as $\kappa\rightarrow\infty$ and, for a finite value of κ, the difference between $\epsilon_n(\kappa)$ and $\epsilon_n(\infty)$ represents the effect of final-state interactions neglected in the IA. The coefficients $\epsilon_n(\kappa)$ were evaluated for $n=0, 1, 2, 3$, and 4, ignoring interference effects in the scattering process. They lead to the conclusion that final-state interactions were not negligible over the range of κ values (2–9 Å^{-1}) employed in the recent experiments of Cowley and Woods.[2]

The interference effects ignored in I[1] are investigated in the present paper. It is shown in Sec. II that when interference effects are included, $\epsilon_n(\kappa)$ can still be calculated exactly for $n=0$ and 1. Approximate expressions for the coefficients with $n=2, 3$, and 4 are proposed and shown to (i) reduce to the correct incoherent expressions as $\kappa\rightarrow\infty$ and (ii) when expanded in powers of $\hbar^2$ yield not only the correct classical limits but (when $n=2$ or 3) also give the correct expressions for the first quantum corrections. The scattering function obtained by truncating the Gram-Charlier series after the $n=4$ term is found to be in reasonable agreement with the data of Cowley and Woods for $T=1.1\,^\circ\text{K}$ and $4<\kappa<9\,\text{Å}^{-1}$.

The temperature dependence of the scattering function is considered in Sec. III. The main problem here is the calculation of the rms velocity of a He^4 atom. It is shown that this quantity can be expressed approximately in terms of its value at $T=0$ and the thermodynamic properties of the liquid. Again satisfactory agreement with the data of Cowley and Woods is obtained.

Finally, in Sec. IV a few concluding remarks are presented.

II. INTERFERENCE EFFECTS

The first-two central moments[1] of the coherent scattering function are given by[3]

$$s_0(\kappa)=1+\gamma(\kappa),\quad s_1(\kappa)=-\omega_r\gamma(\kappa)\,, \tag{1}$$

where $\omega_r=\hbar\kappa^2/2m$ is the recoil energy and

$$\gamma(\kappa)\equiv S(\kappa)-1=(N/\Omega)\int\cos\vec{\kappa}\cdot\vec{r}\,[g(r)-1]\,d\vec{r}. \tag{2}$$

$s_0(\kappa)$ and $s_1(\kappa)$ are uniquely determined by the pair correlation function $g(r)$. However, the higher-order moments are not, because, in general, they involve averages over the positions of three or more atoms as well as simultaneous averages over positions and velocities. These latter quantities are not statistically independent when quantum effects are important, as in the case in liquid helium. The situation is somewhat simpler at temperatures high enough for quantum effects to be small, where the following expansions in powers of $\hbar^2$ may be used[4]:

$$s_2(\kappa)=\kappa^2\frac{kT}{m}+\hbar^2\left(\frac{\kappa^4\gamma_{cl}(\kappa)}{4m^2}+\frac{\kappa^2B_{cl}(\kappa)}{12mkT}\right)+O(\hbar^4)\,,$$

$$s_3(\kappa)=\hbar\left(\frac{\kappa^2B_{cl}(\kappa)}{2m}\right)+O(\hbar^3)\,, \tag{3}$$

$$s_4(\kappa)=3\kappa^4\left(\frac{kT}{m}\right)^2+\kappa^2\frac{kT}{m}B_{cl}(\kappa)+O(\hbar^2)\,.$$

Here $\gamma_{cl}(\kappa)$ denotes the classical limit of $\gamma(\kappa)$, and $B_{cl}(\kappa)$ the classical limit of

$$B(\kappa) \equiv \frac{N}{m\Omega} \int (1 - \cos\kappa x)\, \frac{\partial^2 \phi(r)}{\partial x^2}\, g(r)\, d\vec{r}, \qquad (4)$$

where the x axis is taken in the direction of $\vec{\kappa}$. For sufficiently large values of κ, on the other hand, interference effects disappear and[5]

$$s_2(\kappa) = \kappa^2 \langle v_\kappa^2 \rangle \ , \qquad s_3(\kappa) = \omega_r\, B(\infty)\ ,$$
$$s_4(\kappa) = \kappa^4 \langle v_\kappa^4 \rangle + \kappa^2 \langle v_\kappa^2 \rangle\, B(\infty)\ . \qquad (5)$$

Since[3] $$\langle v_\kappa^2 \rangle = \frac{kT}{m} + \frac{\hbar^2 B_{\text{cl}}(\infty)}{12mkT} + O(\hbar^4)\ , \qquad (6)$$

the simplest expressions satisfying both (3) and (5) are evidently

$$s_2(\kappa) = \kappa^2 \langle v_\kappa^2 \rangle + \omega_r^2 \left(\gamma(\kappa) + \frac{B(\kappa) - B(\infty)}{3\kappa^2 \langle v_\kappa^2 \rangle} \right),$$
$$s_3(\kappa) = \omega_r\, B(\kappa)\ , \qquad (7)$$
$$s_4(\kappa) = \kappa^4 \langle v_\kappa^4 \rangle + \kappa^2 \langle v_\kappa^2 \rangle\, B(\kappa)\ .$$

These approximate relations can be regarded as partial summations of the series expansions in powers of $\hbar^2$ given by Eq. (3). The incoherent terms have been summed exactly, but only those interference terms arising from quantum effects in the pair correlation function have been included.

Substituting (1) and (7) into (I 22), and taking $\alpha = \kappa(\frac{1}{2}\langle v_\kappa^2 \rangle)^{1/2}$ as before, one obtains, for the coefficients in the Gram-Charlier expansion,

$$\epsilon_0(\kappa) = 1 + \gamma(\kappa)\ ,$$
$$\epsilon_1(\kappa) = -(\kappa/\kappa_1)\gamma(\kappa)\ ,$$
$$\epsilon_2(\kappa) = \{\tfrac{1}{2}(\kappa/\kappa_1)^2 - 1\}\, \gamma(\kappa) + (\kappa_3/2\kappa_1)\, \mu(\kappa)\ , \qquad (8)$$
$$\epsilon_3(\kappa) = (\kappa/\kappa_1)\gamma(\kappa) + (\kappa_3/\kappa)\{1 + \mu(\kappa)\},$$
$$\epsilon_4(\kappa) = \zeta_4 + (\kappa_4/\kappa)^2 + \tfrac{1}{2}[1 - (\kappa/\kappa_1)^2]\gamma(\kappa)$$
$$\qquad + [(\kappa_4/\kappa)^2 - (\kappa_4/\kappa_1)^2]\, \mu(\kappa)\ ,$$

where $\hbar\kappa_1 = m(2\langle v_\kappa^2 \rangle)^{1/2}$, $\mu(\kappa) = B(\kappa)/B(\infty) - 1$, (9)

and the remaining parameters κ_3, κ_4, and ζ_4 are given by (I 30) and (I 38). The parameters in (8) can be evaluated at $T = 0$ with the help of the pair correlation function and velocity distribution function computed by McMillan[6] from a variational ground-state wave function of the Jastrow type. It was found in I that $\langle v_\kappa^2 \rangle = 1.96 \times 10^8$ cm^2 s^{-2}, $\zeta_4 = 0.092$, $\kappa_3 = 4.2$ Å^{-1}, and $\kappa_4 = 1.6$ Å^{-1}. Hence, $\kappa_1 = 1.3$ Å^{-1}. McMillan's pair correlation function has also been used to calculate $\gamma(\kappa)$ and $\mu(\kappa)$ numerically, with results shown in Fig. 1, together with the corresponding $\epsilon_n(\kappa)$. It is seen that $\mu(\kappa)$, and hence $\epsilon_2(\kappa)$, $\epsilon_3(\kappa)$, and $\epsilon_4(\kappa)$ are much more strongly oscillating functions of κ than is $\gamma(\kappa)$. Consequently, the shape of the scattering function is much more sensitive to interference effects than is its integrated intensity.

The Gram-Charlier expansion (I 20) for the scattering function is

$$S(\kappa, \omega) = \frac{1}{2\sqrt{\pi}\alpha} \exp\left[- \left(\frac{\omega - \omega_r}{2\alpha} \right)^2 \right]$$
$$\times \sum_{n=0}^{\infty} \epsilon_n(\kappa)\, H_n\left(\frac{\omega - \omega_r}{2\alpha} \right)\ . \qquad (10)$$

Since $\epsilon_n(\kappa)$ is determined according to (I 22) by the moments $s_m(\kappa)$ for $m = n$, $n-2$, $n-4$, ..., the series (10) has the property that if it is truncated after the nth term, the truncated $S(\kappa, \omega)$ differs from the complete $S(\kappa, \omega)$ only insofar as moments of order greater than n are concerned. Thus, if expressions (8) are employed in (10) and the series is truncated after the term $n = 4$, the truncated $S(\kappa, \omega)$ satisfies the $m = 0$ and 1 moment relations identically, and has approximately the correct

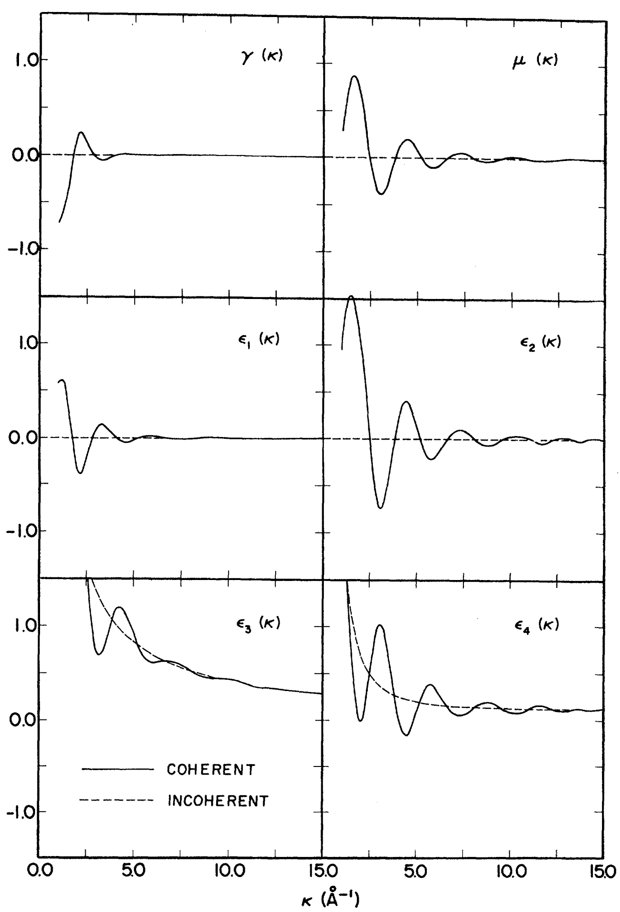

FIG. 1. Coefficients in the Gram-Charlier series (10) calculated for $T = 0$ from the expressions (8) with the help of McMillan's pair correlation function and velocity distribution function. The dashed lines represent the corresponding incoherent coefficients which are obtained from (8) by putting $\gamma(\kappa)$ and $\mu(\kappa)$ equal to their asymptotic values, zero, in the limit $\kappa \to \infty$.

$m = 2$, 3 and 4 moments for all κ.

The truncated series was used to solve numerically the equations

$$\frac{\partial S(\kappa, \omega)}{\partial \omega} = 0, \quad \text{when } \omega = \omega_{\max}$$
$$S(\kappa, \omega) = \tfrac{1}{2} S(\kappa, \omega_{\max}), \quad \text{when } \omega = \omega_{\pm} . \tag{11}$$

The solutions determine the full width at half-maximum

$$\Delta\omega = \omega_+ - \omega_- , \tag{12}$$

and the mean frequency

$$\omega_m = \tfrac{1}{2}(\omega_+ + \omega_-) . \tag{13}$$

If $S(\kappa, \omega)$ were symmetrical about the maximum, ω_m would equal $\omega_{\max}$. $S(\kappa, \omega)$ is not symmetrical. Nevertheless, these quantities differ by less than 1% when $\kappa > 4$ Å^{-1} because the asymmetry is confined almost entirely to the wings of the scattering function.

The calculated quantities $\Delta\omega$ and ω_m are shown in Figs. 2 and 3, together with the data measured by Cowley and Woods[2] at 1.1 °K. General qualitative agreement between the observed and calculated curves exists although significant quantitative discrepancies occur. These discrepancies can be attributed to a number of causes:

(i) The coefficients $\epsilon_n(\kappa)$ were calculated with the help of McMillan's approximate wave function which gives[6] a ground-state energy 20% above the experimental value and also gives a pair correlation function which differs by roughly 20% from those obtained from x-ray and neutron-diffraction experiments. Moreover, the oscillations in $g(r)$ and $\gamma(\kappa)$ found by McMillan are not as pronounced

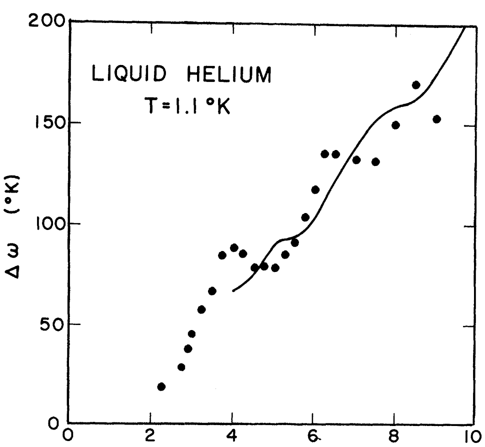

FIG. 2. Comparison of observed and calculated halfwidths. The full line is the curve calculated from the coefficients shown in Fig. 1. The dots represent the observed data of Cowley and Woods.[2]

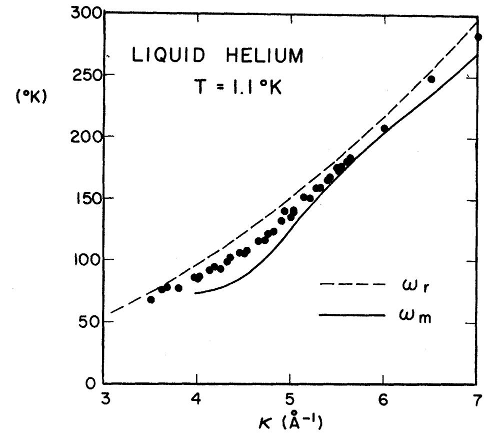

FIG. 3. Comparison of observed and calculated mean frequencies. The full line is the curve calculated from the coefficients shown in Fig. 1. The dots represent (unpublished) data of Cowley and Woods. The dashed line is the free-particle recoil energy $\omega_r = \hbar\kappa^2/2m$.

as is observed experimentally. Quite apart from other sources of error it is, therefore, not surprising that one finds discrepancies between the observed and calculated $\Delta\omega$, which are typically 20%, and that the amplitude of the observed oscillation is larger than is calculated.

(ii) The approximate nature of expressions (8) for $\epsilon_2(\kappa)$, $\epsilon_3(\kappa)$, and $\epsilon_4(\kappa)$ will produce errors in $\Delta\omega$ and ω_m especially for small κ, where interference effects are largest.

(iii) It is difficult to assess the error introduced by truncating the Gram-Charlier series. Since the truncation changes only the fifth- and higher-order moments of $S(\kappa, \omega)$, one is tempted to conclude that the truncation error is important only in the wings of $S(\kappa, \omega)$ and, hence, will have little effect on the values of $\Delta\omega$ and ω_m. However, this is certainly not true for values of κ large enough for the impulse approximation to be valid since truncating (I 32) will tend to suppress the zero-momentum peak.

(iv) The fact that the curves in Figs. 2 and 3 are calculated for $T = 0$ while the data were taken at a finite temperature of 1.1 °K is a further source of error although the results of Sec. III suggest that this error is quite negligible.

In view of the above remarks, the agreement between theory and experiment in Figs. 2 and 3 can be regarded as satisfactory. In particular, it seems reasonable to assert that the oscillations in the observed $\Delta\omega$ data arise from interference effects which, in turn, are mainly a reflection of the hard core of the interatomic potential. We emphasize this point because Cowley and Woods[2] had originally assumed that the observed oscillations did not arise from interference effects [because $\gamma(\kappa)$

is essentially zero when $\kappa > 4\,\text{Å}^{-1}$] and that, therefore, one must search elsewhere for an explanation. They speculated that the oscillations might arise from the existence of thresholds above which a recoiling He^4 atom can create small quantized vortex rings having successively 1, 2, . . . , units of vorticity. However, this novel suggestion is, we feel, weakened by the fact that the oscillations are observed[7] to persist with roughly the same amplitude above the λ point where liquid helium behaves like a Newtonian liquid with unquantized vorticity. We consider it more likely that the oscillations in $\Delta\omega$ are due entirely to interference effects and should, therefore, be regarded as analogous to the de Gennes narrowing effect[4] in classical liquids.

Fernandez and Gersh[8] suggested that the observed oscillations in $\Delta\omega$ arise from the oscillations in $\gamma(\kappa)$. This suggestion is based on the assumption that $\gamma(\kappa)$ has the form $(\cos\kappa\sigma)/\kappa^2$ appropriate for a dilute classical hard-sphere gas. However, this assumption is not valid for liquid helium. A realistic model for $g(r)$ will go smoothly to zero as $r \to 0$ with the result that the oscillations in $\gamma(\kappa)$ will damp out more rapidly than κ^{-2}. This is clear from Fig. 1, where it is seen that McMillan's pair correlation function indicates that $\gamma(\kappa)$ is virtually zero above $5\,\text{Å}^{-1}$.

We now consider the shape of the scattering function. For this purpose it is convenient to express $\Delta\omega$ in the form

$$\Delta\omega = \Delta\omega_g (1-\chi), \tag{14}$$

where $\Delta\omega_g$ is the full width of $S(\kappa,\omega)$ at half-maximum for the Gaussian approximation in which $\epsilon_n(\kappa)$ is put equal to zero when $n > 0$:

$$\Delta\omega_g = \kappa (8 \ln 2 \langle v_\kappa^2 \rangle)^{1/2}, \tag{15}$$

and χ describes the narrowing due to the non-Gaussian terms in the Gram-Charlier series. The effect of the shape of the scattering function on $\Delta\omega$ is contained entirely in χ, and $\Delta\omega_g$ plays the role of a scaling factor.

The truncated Gram-Charlier series gives a scattering function which is approximately Gaussian near the center, but which departs asymmetrically from a Gaussian in the wings. The high-frequency wing is enhanced and the low-frequency wing depressed. The asymmetry decreases with increasing κ. The neutron-data also shows this behavior[7] although the data is not sufficiently precise to determine the shape of the wings accurately.

If one assumes a Gaussian line shape by putting $\Delta\omega = \Delta\omega_g$ and then fits a straight line through the origin to the data in Fig. 2, one finds an effective value for $\langle v_\kappa^2\rangle$ equal to $1.13\times10^8\ \text{cm}^2\,\text{sec}^{-2}$. This is 73% smaller than McMillan's value[8] of $1.96\times10^8\ \text{cm}^2\,\text{sec}^{-2}$. On the other hand, it is seen in Fig. 2 that McMillan's value gives quite reasonable agreement when the non-Gaussian terms up to $n=4$ are included. While McMillan's value may perhaps be in error by as much as 20%, we consider it unlikely to be in error by 73%. Thus, we conclude that the scattering function for liquid He^4 at 1.1°K does depart significantly from a Gaussian line shape in the wings.

III. TEMPERATURE DEPENDENCE OF SCATTERING FUNCTION

To determine the temperature dependence of the scattering function we first consider $\langle v_\kappa^2\rangle$ and note that this quantity occurs in the general expressions for both the thermal and caloric equations of state

$$\begin{aligned} pv &= \langle v_\kappa^2\rangle - \frac{2\pi\rho}{3m^2}\int_0^\infty \frac{d\phi(r)}{dr}\, g(r)\, r^3\, dr, \\ u &= \tfrac{3}{2}\langle v_\kappa^2\rangle + \frac{2\pi\rho}{m^2}\int_0^\infty \phi(r)\, g(r)\, r^2 dr, \end{aligned} \tag{16}$$

where p is the pressure, v the volume per unit mass, $\rho = 1/v$ the density, and u the internal energy per unit mass.

Henshaw[9] has measured the structure factor $\gamma(\kappa)$ both above and below the λ point and found that for values of κ above the position of the first diffraction maximum $\gamma(\kappa)$ is independent of temperature. This implies that $g(r)$ varies appreciably with temperature only for large values of r. The pair potential is proportional to r^{-6} when r is large so that one can write

$$r\frac{d\phi(r)}{dr} \doteq -6\phi(r) - \lambda(r), \tag{17}$$

in which the quantity $\lambda(r)$ so defined is a short-range[10] function of r. Combining (16) and (17), we obtain

$$\langle v_\kappa^2\rangle = \Lambda + u - \tfrac{1}{2}pv, \tag{18}$$

where $$\Lambda = (\pi\rho/3m^2)\int_0^\infty \lambda(r)\, g(r)\, r^2\, dr. \tag{19}$$

In what follows, we shall adopt the convention that quantities labeled with the subscript 0 refer to the state $T=0$, $p=0$; while unlabeled quantities refer to an arbitrary state of the liquid in equilibrium under its own saturated vapor pressure. Thus,

$$\langle v_\kappa^2\rangle_0 = \Lambda_0 + u_0. \tag{20}$$

Since $\lambda(r)$ is a short-range function of r, the integrand of (19) is appreciably different from zero only for small values of r, where Henshaw's experiments indicate $g(r)$ is independent of temperature. Hence,

$$\frac{\Lambda}{\rho} = \frac{\Lambda_0}{\rho_0}. \tag{21}$$

Combining (18), (20), and (21) we get

$$\langle v_\kappa^2\rangle = (\rho/\rho_0)\langle v_\kappa^2\rangle_0 + (1-\rho/\rho_0)u_0 + \Delta u - \tfrac{1}{2}pv, \quad (22)$$

where $\Delta u \equiv u - u_0$ is the change in internal energy, which is given by the first law of thermodynamics:

$$\Delta u = \int_0^T C dT - \int_{v(0)}^{v(T)} p\, dv, \quad (23)$$

in which C is the orthobaric specific heat per unit mass. Thus, Eq. (22) expresses $\langle v_\kappa^2\rangle$ in terms of its value at $T=0$ and the known thermodynamic properties of liquid helium.[11] If McMillan's value $\langle v_\kappa^2\rangle_0 = 1.96\times 10^8$ cm^2 s^{-2} is used in (22), one finds that $\langle v_\kappa^2\rangle = 2.53\times 10^8$ cm^2 s^{-2} at the boiling point (4.2 °K). This corresponds to an increase in the average kinetic energy per atom of 4.1 °K and is consistent with the value 3°K which Puff and Tenn[12] speculate might be reasonable.

Below 2.5°K, thermal expansion is negligible and the vapor pressure is so small that (22) reduces to

$$\langle v_\kappa^2\rangle \simeq \langle v_\kappa^2\rangle_0 + \int_0^T C\, dT. \quad (24)$$

Above 2.5 °K, however, p increases rapidly and ρ decreases with the result that $\langle v_\kappa^2\rangle$ increases less rapidly than the specific-heat integral.

The temperature dependence of the parameter α in the Gram-Charlier series (10) is determined entirely by $\langle v_\kappa^2\rangle$, while the temperature dependence of $\epsilon_n(\kappa)$ is determined by both $\langle v_\kappa^2\rangle$[through the quantities κ_1, κ_3, and κ_4 in Eq. (8)] and ρ. For large values of κ, the integral in the expression (4) for $B(\kappa)$ will, like that for $\gamma(\kappa)$, be independent of temperature. For the remaining parameter ζ_4 in Eq. (8) we shall assume that $\langle v_\kappa^4\rangle$ has the same temperature dependence as $\langle v_\kappa^2\rangle^2$ so that ζ_4 is independent of T.

It may be seen from Eq. (8) that, generally speaking, the magnitude of $\epsilon_n(\kappa)$ for $n>0$ decreases as $\langle v_\kappa^2\rangle$ increases and increases as ρ increases. As the temperature is decreased below the λ point, ρ remains constant and $\langle v_\kappa^2\rangle$ decreases, so that $\epsilon_n(\kappa)$ increases and the line shape becomes less Gaussian. On the other hand, as the temperature is increased above the λ point, $\langle v_\kappa^2\rangle$ increases and ρ decreases, with the result that $\epsilon_n(\kappa)$ decreases and the line shape becomes more Gaussian. These qualitative conclusions are confirmed by the numerical calculations discussed below.

$\Delta\omega$ has been calculated as a function of T for $\kappa = 5.1$ Å^{-1} following the method outlined in Sec. II, and the results are shown in Fig. 4 together with the data of Cowley and Woods.[2] As the temperature is decreased from 4.2°K to 0°K, the calculated value of $\Delta\omega$ decreases from 109 to 91 °K, while $\Delta\omega_g$, defined by Eq. (15), decreases from

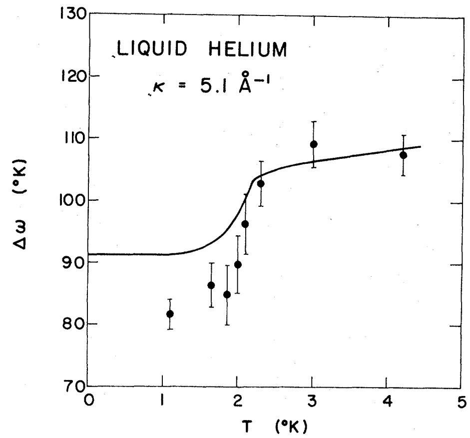

FIG. 4. Comparison of observed and calculated half-widths as functions of temperature. The full line is the curve calculated with the help of the approximate relation (22). The dots represent the observed data of Cowley and Woods.[2]

146 to 129 °K. This corresponds to an increase in χ from 0.25 to 0.29. If χ were constant, i.e., if the coefficients $\epsilon_n(\kappa)$ were temperature independent, the decrease in $\Delta\omega$ would merely describe a change in the width of $S(\kappa,\omega)$ without any change in shape. The fact that χ is not constant, but increases with decreasing temperature, shows that $S(\kappa,\omega)$ is less Gaussian and more strongly peaked below the λ point than it is above.

Up to this point the temperature dependence of the scattering has been discussed in terms of the observed thermodynamic properties of liquid helium. These properties are, in turn, determined by the Bose condensation. It is of interest to try to interpret the above results in microscopic terms.

Equation (24) is clearly consistent with the generally accepted view that the λ anomaly in the specific heat of liquid He^4 is due to a Bose condensation. In particular the decrease in $\langle v_\kappa^2\rangle$ as the temperature is decreased below the λ point can be interpreted by noting that the finite fraction of atoms in the condensate gives a vanishing contribution to $\langle v_\kappa^2\rangle$, so that as this fraction increases below the λ point $\langle v_\kappa^2\rangle$ shows a corresponding decrease.

The slight peaking of $S(\kappa,\omega)$ below the λ point can perhaps be interpreted in terms of the appearance of an unresolved zero-momentum peak. The zero-momentum peak has been predicted[1] to become a prominent feature of $S(\kappa,\omega)$ in the limit $\kappa\to\infty$. It was pointed out in Sec. II that the truncation of the Gram-Charlier series would tend to suppress the zero-momentum peak. This may account for the nature of the discrepancy between the observed and calculated widths in Fig. 4. Above

T_λ, where there is no zero-momentum peak, the calculated width is in good agreement with the neutron data. Below T_λ the calculated width does not fall to as low a value as the observed width, owing to a partial suppression of the zero-momentum peak caused by truncating the Gram-Charlier series. It must be emphasized that this interpretation is purely speculative since the widths in Fig. 4 are for $\kappa = 5.1\,\text{Å}^{-1}$, where final-state interactions are large and the concept of a zero-momentum peak is ill defined.

Finally, we point out that the fact that $\Delta\omega$ is constant up to approximately $1.5\,°\text{K}$ justifies our having compared data taken at $1.1\,°\text{K}$ in Figs. 2 and 3 with calculations for $T = 0$.

IV. CONCLUDING REMARKS

The most interesting problem remaining to be solved concerns the zero-momentum peak and, more generally, the way in which the scattering function approaches its asymptotic limit, the impulse approximation, as $\kappa \to \infty$. It is felt that no reliable information on this question will be obtained merely by calculating a few more terms in the Gram-Charlier series. Rather, one must determine how $\epsilon_n(\kappa)$ approaches ζ_n as $\kappa \to \infty$ for arbitrary n. In other words, the first few terms in the Gram-Charlier series are determined by the low-order moments of $S(\kappa, \omega)$ which, in turn, are determined by the behavior of $F(\kappa, t)$ at small t. The shape of the zero-momentum peak, on the other hand, is determined by the long-time behavior of $F(\kappa, t)$. For this reason, attempts[12, 13] to understand the zero-momentum peak by using low-order moment relations to determine the parameters in an *ad hoc* model for $S(\kappa, \omega)$ are liable to yield misleading results.

ACKNOWLEDGMENTS

Discussions with Dr. A. D. B. Woods, Dr. R. A. Cowley, and Professor L. J. Sham are gratefully acknowledged.

---

[1]V. F. Sears, Phys. Rev. 185, 200 (1969), referred to as I. The notation and terminology introduced in I will be followed throughout the present article. Equations in I will be referred to by the appropriate number prefaced by the numeral I.

[2]R. A. Cowley and A. D. B. Woods, Phys. Rev. Letters 21, 787 (1968); *Neutron Inelastic Scattering* (International Atomic Energy Agency, Vienna, 1968), Vol. I, p. 609.

[3]A. Rahman, K. S. Singwi, and A. Sjölander, Phys. Rev. 126, 986 (1962).

[4]The classical limits of the moments have been derived by de Gennes [Physica 25, 825 (1959)]. The quantum corrections can be obtained either by the method suggested in Ref. 3 or, more directly, by extending to coherent scattering the method based on the principle of detailed balance, which is discussed by Nelkin [*Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1961), p. 3] for incoherent scattering.

[5]To compare the expressions (5) with those given in I note that $B(\infty) = \langle \Delta V \rangle / 3m$. In the expression for $s_4(\kappa)$, we employ the approximation (I C8).

[6]W. L. McMillan, Phys. Rev. 138, A442 (1965).

[7]R. A. Cowley and A. D. B. Woods (private communication).

[8]J. F. Fernandez and H. A. Gersch, Phys. Letters 30A, 261 (1969).

[9]D. G. Henshaw, Phys. Rev. 119, 9 (1960).

[10]For the Lennard-Jones potential $\lambda(r) \sim r^{-12}$.

[11]The calculations which follow are based on data taken from the following sources: C below T_λ is from H. C. Kramers, J. D. Wasscher, and C. J. Gorter, Physica 18, 329 (1952); C above T_λ from R. W. Hill and O. V. Lounasmaa, Phil. Mag. 2, 143 (1957); the logarithmic singularity in C at T_λ was integrated over analytically with the help of the interpolation formula of M. J. Buckingham and W. M. Fairbank [Progr. Low Temp. Phys. 3, 80 (1961)]; ρ is from E. C. Kerr [J. Chem. Phys. 26, 511 (1957)]; p is from H. Van Dijk and M. Durieux [Progr. Low Temp. Phys. 2, 431 (1957)], and, finally, the observed value of u_0 and the calculated value of $\langle v_\kappa^2 \rangle_0$ given by McMillan (Ref. 6) were used.

[12]R. D. Puff and J. S. Tenn (unpublished).

[13]P. A. Egelstaff and R. D. Mountain (unpublished).