Neutron Scattering from Liquid Helium II at Large Momentum Transfer and the Condensate Fraction*

W. C. Kerr†
Argonne National Laboratory, Argonne, Illinois 60439
and
K. N. Pathak‡
Department of Physics, Northwestern University, Evanston, Illinois 60201
and
K. S. Singwi
Department of Physics, Northwestern University, Evanston, Illinois 60201
and Argonne National Laboratory, Argonne, Illinois 60439
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A simple theory of neutron scattering from liquid He II at zero temperature for large momentum transfers is described. The theory is based on a generalized mean-field approximation involving the polarization potential and the screened response function. The latter, instead of having a free-particle form, is assumed to be a sum of Gaussian functions, weighted by the momentum distribution function appropriate for liquid helium. The zero and third moments of the scattering law determine the polarization potential and the width of the Gaussians. Numerical calculations have been made for the dynamical structure function $S(q, \omega)$ for different values of the condensate fraction. Both the width and the peak position of $S(q, \omega)$ in the range of momentum transfers 2.5–9 Å^{-1} are in fair agreement with experiment for a 6% condensate fraction. Calculations have also been done for q values as large as 20 Å^{-1} for three different values of the condensate fraction, including 0. For a condensate fraction of 6% or larger, the contribution of the condensate to $S(q, \omega)$ becomes separated from that of the noncondensed particles at these large-q values. The separation is distinct enough that it should be experimentally observable.

I. INTRODUCTION

The microscopic basis for the unusual properties of liquid helium is generally believed to be the occupation of the zero-momentum quantum state by a macroscopic number of particles. The original proposal of this idea, by London,[1] was based on analogy with the Bose-Einstein condensation which occurs for the free Bose gas. Onsager and Penrose[2] gave the proper mathematical formulation of this concept for a system of interacting particles, and made the first estimate of the fraction of particles which are in the condensed state.

The experimental evidence for this so-called condensate is mainly of an indirect nature. It has been recognized for some time that neutron scattering experiments could provide more direct experimental evidence concerning the existence of the condensate.[3] A proposal for an experiment specifically designed to probe this problem was made by Hohenberg and Platzman.[4] They suggested that if neutrons were scattered from liquid helium at sufficiently high momentum and energy transfers the response of the helium atoms could be treated in a nearly free-particle approximation. The existence of a condensate could then show up in the distribution of scattered neutrons as a more or less sharp peak centered at the free-particle recoil energy, because the condensate provides a large number of particles in the zero-momentum state.

Cowley and Woods[5] performed an experiment to test these ideas using momentum transfers in the range 2.5–9 Å^{-1} ($\times\hbar$). The scattering law obtained by them, as a function of energy transfer for fixed momentum transfer, has a single smoothly shaped peak in it, and it can be characterized by the position of the maximum and the width at half-maximum. The sharp peak expected from Hohenberg and Platzman's argument does not seem to be present. Thus, the experiment does not provide the direct verification of the existence of a condensate that was hoped for, but it is not inconsistent with there being a condensate.

There are two interesting features of the results obtained by Cowley and Woods. First, the position of the maximum in the cross section is not quite at the free-particle recoil energy but is shifted to lower energies. Second, the width of the peak has definite oscillations as a function of the momentum transfer.

In this paper we propose a simple theory to account for Cowley and Woods's experimental results. Our theory is based on a mean-field approach to calculating the cross section and the requirement that the low-order sum rules for the cross section be exactly satisfied. We find both a shift in the maximum of the cross section and structure in the width, and our results are in fair agreement with the experimental results. These features of the

cross section result from structure in both the static structure factor and the third frequency moment of the cross section. The fraction of particles in the condensed state is the one parameter in our theory, and the value we obtain for that is consistent with other recent estimates.[6]

In addition to comparing our calculations with the results of Cowley and Woods's experiment, we present calculations for values of momentum transfer which are considerably larger than those used in their experiment. We find that for sufficiently large momentum transfer the cross section takes on the qualitative shape proposed by Hohenberg and Platzman, namely, a distinct peak due to the condensate sitting on a broad background due to the rest of the particles.

The outline of this paper is as follows. Section II contains the formal discussion of the scattering law, the sum rules, and the details of our model. The numerical results are presented in Sec. III, and the conclusions are summarized in Sec. IV. Some computational details are discussed in the Appendix.

II. FORMULATION

A. Generalities

The function which is important in the theory is the density response function $\chi(q,\omega)$. The general properties of this function have been discussed in the literature,[7] so the description here is very brief.

If an infinitesimal external disturbance which couples to the density is imposed on the system, the Fourier components of the induced density change are related to the Fourier components of the external disturbance by

$$\delta\langle\rho(\vec{q},\omega)\rangle=\chi(q,\omega)\,V_{\text{ext}}(\vec{q},\omega). \tag{2.1}$$

From standard linear response theory,[8] the density response function in space and time variables is

$$\chi(\vec{x},t)=(-i/\hbar)\,\theta(t)\langle[\rho(\vec{x},t),\ \rho(\vec{O},O)]\rangle, \tag{2.2}$$

where

$$\rho(\vec{x},t)=\sum_{i=1}^{N}\delta(\vec{x}-\vec{x}_i(t)),$$

is the density operator, and $\theta(t)$ is the unit step function, being 0 for $t<0$ and 1 for $t>0$.

Since $\chi(\vec{x},t)$ is a retarded response function, $\chi(q,\omega)$ is analytic in the upper half of the complex ω plane, and its real and imaginary parts, denoted by $\chi'(q,\omega)$ and $\chi''(q,\omega)$, respectively, are related by the Kramers-Kronig relations.[9] $\chi''(q,\omega)$ is an odd function of ω and $\omega\chi''(q,\omega)\leq 0$. Furthermore, $\chi''(q,\omega)$ is the spectral function for $\chi(q,\omega)$, i.e.,

$$\chi(q,\omega)=(1/\pi)\int_{-\infty}^{\infty}d\omega'[\chi''(q,\omega')/(\omega'-\omega)] \tag{2.3}$$

for ω in the upper-half plane.

From the spectral representation Eq. (2.3) the large-ω expansion of $\chi(q,\omega)$ is obtained as

$$\chi(q,\omega)=-(1/\pi\omega^2)\int_{-\infty}^{\infty}d\omega'\,\omega'\chi''(q,\omega')-(1/\pi\omega^4)\int_{-\infty}^{\infty}d\omega'\omega'^3\chi''(q,\omega')\ldots. \tag{2.4}$$

The coefficients in the asymptotic expansion of $\chi(q,\omega)$ are the odd frequency moments of $\chi''(q,\omega)$.

The experimentally measured function is the scattering law or dynamical structure function[20] $S(q,\omega)$. The Fourier transform of $(2\pi)S(q,\omega)$ is the density correlation function

$$G(\vec{x},t)=n^{-1}\langle\rho(\vec{x},t)\,\rho(\vec{O},O)\rangle, \tag{2.5}$$

where n is the average density. The fluctuation-dissipation theorem[11] relates $\chi''(q,\omega)$ to $S(q,\omega)$ as

$$S(q,\omega)=-(\hbar/\pi n)(1-e^{-\beta\hbar\omega})^{-1}\chi''(q,\omega), \tag{2.6}$$

where β is the inverse temperature.

The low-order moment relations or sum rules for $S(q,\omega)$ are well known.[12] The integral of $S(q,\omega)$ is the static structure factor $S(q)$, and the first frequency moment is the f sum rule. The third frequency moment has been derived by Puff.[13,14] These sum rules can be stated in terms of $\chi''(q,\omega)$, using Eq. (2.6), as

$$-(1/\pi)\int_{-\infty}^{\infty}d\omega\,\chi''(q,\omega)\coth\tfrac{1}{2}\beta\hbar\omega=(2n/\hbar)S(q), \tag{2.7}$$

$$-(1/\pi)\int_{-\infty}^{\infty}d\omega\,\omega\chi''(q,\omega)=(n/M)q^2, \tag{2.8}$$

$$-(1/\pi)\int_{-\infty}^{\infty}d\omega\,\omega^3\chi''(q,\omega)=(n/M^2)q^4\left[2\langle\text{KE}\rangle/N+\hbar^2q^2/4M+n(1/q^2)\times\int d\vec{x}\,g(x)(1-\cos\vec{q}\cdot\vec{x})(\hat{q}\cdot\nabla)^2V(x)\right]. \tag{2.9}$$

Here $\langle\text{KE}\rangle/N$ denotes the average kinetic energy per particle, $g(x)$ is the static pair correlation function, which is related to $S(q)$ by

$$n[g(x)-1]=(2\pi)^{-3}\int d\vec{q}\,e^{i\vec{q}\cdot\vec{x}}[S(q)-1],$$

and $V(x)$ is the interparticle potential.

Equations (2.8) and (2.9) express the coefficients in the asymptotic expansion of $\chi(q,\omega)$ in terms of the interparticle potential and the static pair correlation function.

The results in Eqs. (2.7)–(2.9) are obtained from the coefficients in the short-time expansion of $\chi(\vec{x},t)$. Thus these sum rules give information on the motion of the particles over short-time intervals only. For example, the first-moment sum rule can be interpreted as saying that over a sufficiently short-time interval, the particles move

as free particles. The effects of the interparticle potential on the dynamics first enter in the third frequency moment. The high momentum and energy transfer experiment of Cowley and Woods probes the motion of the particles over short distances and over short-time intervals. Therefore, satisfaction of these low-order moment relations is probably a minimal requirement on any approximate theory of this experiment.

The rest of the paper is concerned only with the zero-temperature limit of the preceding expressions. In that limit Eq. (2.6) reduces to

$$S(q,\omega)=\begin{cases}-(\hbar/\pi n)\chi''(q,\omega), & \omega>0\\ 0, & \omega<0\end{cases} \tag{2.10}$$

and Eq. (2.7) becomes

$$-(1/\pi)\int_{-\infty}^{\infty}d\omega\,\chi''(q,\omega)=(n/\hbar)S(q). \tag{2.11}$$

B. Model Form for $\chi(q,\omega)$

The model form for $\chi(q,\omega)$ is introduced here and related to the general statements in Sec. II A. This particular model form has been discussed before for liquid helium by Pines[15] and for classical liquids by Singwi, Sköld, and Tosi[16] and by Nelkin.[17]

In Sec. II A the density response function was introduced as the proportionality factor relating a weak external potential $V_{\rm ext}(\vec{q},\omega)$ to the induced density disturbance $\delta\langle\rho(\vec{q},\omega)\rangle$. Due to the interparticle interaction, this density disturbance produces a polarization potential $V_{\rm pol}(\vec{q},\omega)$, which can be written

$$V_{\rm pol}(\vec{q},\omega)=\psi(q)\delta\langle\rho(\vec{q},\omega)\rangle\,. \tag{2.12}$$

Equation (2.12) defines $\psi(q)$, with the additional assumption that it has no frequency dependence. This assumption can be equivalently stated that there is a local relation in time between the polarization potential and the density disturbance.

The *screened* density response function $\chi_{sc}(q,\omega)$ is defined to give the response of the density to the *sum* of the external potential and the polarization potential, i.e.,

$$\delta\langle\rho(\vec{q},\omega)\rangle=\chi_{sc}(q,\omega)[V_{\rm ext}(\vec{q},\omega)+V_{\rm pol}(\vec{q},\omega)]. \tag{2.13}$$

Combining Eqs. (2.1), (2.12), and (2.13) gives

$$\chi(q,\omega)=\chi_{sc}(q,\omega)/[1-\psi(q)\chi_{sc}(q,\omega)]. \tag{2.14}$$

So far we have just introduced two new functions, and we need additional physical assumptions to proceed. We obtain these by the following arguments.

In a neutron scattering event where the momentum and energy transferred to the system are large, we can consider that the neutron interacts with essentially a single helium atom and that atom recoils like a nearly free particle. If the scattering system *were* a free Bose gas, the imaginary part of the response function would be[18]

$$\chi''_{\rm free}(q,\omega)=\pi\hbar^{-1}\textstyle\sum_{\vec{p}}n_{\rm free}(p)[\delta(\omega-\omega_{\vec{p}+\vec{q}}+\omega_{\vec{p}})-\delta(\omega+\omega_{\vec{p}+\vec{q}}-\omega_{\vec{p}})]. \tag{2.15}$$

Here $n_{\rm free}(p)$ is the momentum distribution function for the free Bose gas[19] [at zero temperature $n_{\rm free}(p)=n\delta_{\vec{p},\vec{0}}$], and $\hbar\omega_{\vec{q}}=\hbar^2q^2/2M$ is the free-particle energy-momentum relation. $\chi'_{\rm free}(q,\omega)$ is obtained from the Kramers-Kronig transform of Eq. (2.15).

At the energy and momentum transfers of interest here, we believe that we can treat the response of the particles as nearly free, and we expect the interparticle interactions to have the following three effects. First, the momentum distribution function which enters should be that appropriate for real liquid helium, and not the free particle-distribution function. Second, the mean field acting on the particles should tend to organize their behavior into collective oscillations. This collective behavior is described by the denominator in Eq. (2.14); if there are well-defined collective modes, the denominator vanishes at the collective-mode frequencies. At the q values of interest here, we do not expect to find good collective modes, so the denominator should not have a large effect. Third, the remaining residual interactions damp the nearly free-particle recoil of the helium atoms. Since $\chi_{sc}(q,\omega)$ in the numerator of Eq. (2.14) should describe essentially the single-particle aspects of the motion, we expect that $\chi''_{sc}(q,\omega)$ will be similar to $\chi''_{\rm free}(q,\omega)$ in Eq. (2.15), except that the δ functions will be broadened.

On the basis of these arguments, we assume the following form for $\chi''_{sc}(q,\omega)$:

$$\chi''_{sc}(q,\omega)=-\pi\hbar^{-1}\textstyle\sum_{\vec{p}}n(p)\,[\pi\Gamma(q)]^{-1/2}\times\left(e^{-(\omega-\omega_{\vec{p}+\vec{q}}+\omega_{\vec{p}})^2/\Gamma(q)}-e^{-(\omega+\omega_{\vec{p}+\vec{q}}-\omega_{\vec{p}})^2/\Gamma(q)}\right). \tag{2.16}$$

Here $n(p)$ is the momentum distribution function of liquid helium and the δ functions of Eq. (2.15) have been replaced by Gaussians with a width function $\Gamma(q)$. We choose Gaussians rather than Lorentzians because we want to make use of the frequency moments, which do not exist for a Lorentzian. In the limit that $\Gamma(q)\to 0^+$ and $n(p)\to n_{\rm free}(p)$, Eq. (2.16) goes over to Eq. (2.15).

In a more complete treatment the broadening of the δ functions would be dependent not only on the magnitude of q but also would have frequency dependence. The assumption that there is only q dependence is necessary here to make the problem tractable, and it implies some kind of average over the frequency dependence.

The momentum distribution function $n(p)$ appearing in Eq. (2.16) has the form

$$n(p) = n_0 \delta_{\vec{p},\vec{0}} + \tilde{n}(p). \tag{2.17}$$

The singular term comes from the Bose-Einstein condensation in the system, and it is at this point that the existence of a condensate enters into the theory. The strength of the singular term n_0 which is the density of particles in the condensed state is the one adjustable parameter in our theory. The smooth part $\tilde{n}(p)$ has been calculated by McMillan,[20] and we use his results. In Sec. (II D) we briefly describe the properties of $\tilde{n}(p)$.

$\chi'_{sc}(q, \omega)$ is obtained from the Kramers-Kronig transform of Eq. (2.16). The transform of the Gaussians can be explicitly carried out in terms of the error function of purely imaginary argument.

Equations (2.14) and (2.16) complete the description of our model form for the density response function. The resulting expression contains two unknown functions $\psi(q)$ and $\Gamma(q)$ whose determination is described next.[21]

C. Imposition of the Moment Relations

The model form for $\chi(q, \omega)$ described in Sec. II B gives expressions for the low-order frequency moments of $\chi''(q, \omega)$ in terms of the unknown functions $\psi(q)$ and $\Gamma(q)$. Requiring that these moments agree with the exact expressions in Eqs. (2.8), (2.9), and (2.11) determines these functions.

The moments of $\chi''(q, \omega)$ are the coefficients in the asymptotic expansion of $\chi(q, \omega)$, according to Eq. (2.4). These coefficients can be determined from Eq. (2.14) if the asymptotic expansion of $\chi_{sc}(q, \omega)$ is known. The asymptotic expansion of $\chi_{sc}(q, \omega)$ is determined by the frequency moments of $\chi''_{sc}(q, \omega)$, in exact analogy with the expansion of $\chi(q, \omega)$ in Eq. (2.4). The moments of $\chi''_{sc}(q, \omega)$ can be calculated from the specific form assumed for $\chi''_{sc}(q, \omega)$ in Eq. (2.16).

Thus, we have

$$-(1/\pi)\int_{-\infty}^{\infty} d\omega\, \omega \chi''_{sc}(q, \omega) = (n/M) q^2, \tag{2.18}$$

$$-(1/\pi)\int_{-\infty}^{\infty} d\omega\, \omega^3 \chi''_{sc}(q, \omega) = (n/M^2) q^4 [2\langle \mathrm{KE}\rangle/N + \hbar^2 q^2/4M + (3M/2)\Gamma(q)/q^2]. \tag{2.19}$$

The true kinetic energy per particle appears here because the true $n(p)$ function is used in $\chi''_{sc}(q, \omega)$.

Using these results in the asymptotic expansion of Eq. (2.14) gives

$$-(1/\pi)\int_{-\infty}^{\infty} d\omega\, \omega \chi''(q, \omega) = (n/M) q^2 \tag{2.20}$$

$$-(1/\pi)\int_{-\infty}^{\infty} d\omega\, \omega^3 \chi''(q, \omega) = (n/M^2) q^4 [2\langle \mathrm{KE}\rangle/N + \hbar^2 q^2/4M + (3M/2)\Gamma(q)/q^2 + n\psi(q)]. \tag{2.21}$$

Comparing Eqs. (2.20) and (2.21) with the exact expressions in Eqs. (2.8) and (2.9) shows that the first moment is automatically satisfied and that the third moment is satisfied if

$$(3M/2)\Gamma(q)/q^2 + n\psi(q) = n P_3(q), \tag{2.22}$$

where

$$P_3(q) = (1/q^2)\int d\vec{x} g(x)(1 - \cos \vec{q}\cdot\vec{x})(\hat{q}\cdot\nabla)^2 V(x) \tag{2.23}$$

is the potential part of the third moment.

$P_3(q)$ can be calculated using known values of the pair correlation function $g(x)$ and the interparticle potential $V(x)$. This calculation is discussed in the Appendix.

From Eq. (2.14), $\chi''(q, \omega)$ can be expressed in terms of $\chi'_{sc}(q, \omega)$, $\chi''_{sc}(q, \omega)$, and $\psi(q)$ as

$$\chi''(q, \omega) = \frac{\chi''_{sc}(q, \omega)}{[1 - \psi(q)\chi'_{sc}(q, \omega)]^2 + [\psi(q)\chi''_{sc}(q, \omega)]^2}. \tag{2.24}$$

The explicit expressions for $\chi''_{sc}(q, \omega)$ and $\chi'_{sc}(q, \omega)$ using Eq. (2.16), the Kramers-Kronig transform of Eq. (2.16), and Eq. (2.17) are

$$\chi''_{sc}(q, \omega) = -\frac{\pi^{1/2} n_0}{\hbar [\Gamma(q)]^{1/2}}\left(e^{-(\omega-\omega_q)^2/\Gamma(q)} - e^{-(\omega+\omega_q)^2/\Gamma(q)}\right) + \frac{M}{8\pi\hbar^2 q}\int_0^{\infty} dp\, p\, \tilde{n}(p)\left[\mathrm{erf}\left(\frac{\omega - \omega_q - (\hbar pq/M)}{[\Gamma(q)]^{1/2}}\right) - \mathrm{erf}\left(\frac{\omega - \omega_q + (\hbar pq/M)}{[\Gamma(q)]^{1/2}}\right) + \mathrm{erf}\left(\frac{\omega + \omega_q + (\hbar pq/M)}{[\Gamma(q)]^{1/2}}\right) - \mathrm{erf}\left(\frac{\omega + \omega_q - (\hbar pq/M)}{[\Gamma(q)]^{1/2}}\right)\right], \tag{2.25}$$

and

$$\chi'_{sc}(q, \omega) = \frac{\pi^{1/2} n_0}{\hbar[\Gamma(q)]^{1/2}}\left[e^{-(\omega-\omega_q)^2/\Gamma(q)}\,\mathrm{Im\,erf}\left(\frac{i(\omega - \omega_q)}{[\Gamma(q)]^{1/2}}\right) - e^{-(\omega+\omega_q)^2/\Gamma(q)}\,\mathrm{Im\,erf}\left(\frac{i(\omega + \omega_q)}{[\Gamma(q)]^{1/2}}\right)\right] + \frac{1}{4\pi^{3/2}\hbar[\Gamma(q)]^{1/2}}\int_0^{\infty} dp\, p^2 \tilde{n}(p)\int_{-1}^{1} d\mu\left[e^{-[\omega-\omega_q-(\hbar pq/M)\mu]^2/\Gamma(q)}\,\mathrm{Im\,erf}\left(\frac{i(\omega - \omega_q - (\hbar pq/M)\mu)}{[\Gamma(q)]^{1/2}}\right)\right.$$

$$- e^{-[\omega+\omega_q+(\hbar pq/M)\mu]^2/\Gamma(q)} \operatorname{Im}\operatorname{erf}\left(\frac{i(\omega+\omega_q+(\hbar pq/M)\mu)}{[\Gamma(q)]^{1/2}}\right)\Bigg]. \tag{2.26}$$

Equation (2.22) gives one relation between the two functions $\psi(q)$ and $\Gamma(q)$. We use this relation to eliminate $\Gamma(q)$ from our expressions. Then, for each q, we integrate Eq. (2.24) over ω (numerically) with different values of $\psi(q)$ and then choose $\psi(q)$ so that Eq. (2.11) is satisfied. This then completes the determination of $\psi(q)$ and $\Gamma(q)$.

From Eq. (2.10) we obtain $S(q, \omega)$ and from that we determine the position of the maximum and the width at half-maximum for comparison with the experiments of Cowley and Woods. The results are given in Sec. III.

D. Momentum Distribution Function

The exact momentum distribution function $n(p)$ appears in our expression for the imaginary part of the screened response function in Eq. (2.16). Some properties of $n(p)$ are discussed here.

The form of $n(p)$ for liquid helium which results because of the Bose-Einstein condensation has been given in Eq. (2.17). The value of n_0 is not precisely known, but there have been several calculations or estimates of it.[6,20,22-24] Calculations of $\tilde{n}(p)$ have been made by McMillan,[6] and more recently by Francis, Chester, and Reatto.[24] We have used McMillan's results here.

In this calculation we choose n_0 to be our one parameter, which we adjust to fit the experimental results of Cowley and Woods. In particular, when q becomes as large as 9 Å^{-1}, $\psi(q)$ is practically 0, so we choose n_0 to fit the experimental width of $S(q, \omega)$ at $q = 9$ Å^{-1}, with $\psi(q = 9\ \text{Å}^{-1}) = 0$. Then we use that value of n_0 to do the calculations described in Sec. II C for smaller values of q.

There is a normalization relation between n_0 and $\tilde{n}(p)$ which is

$$n_0 + \sum_{\vec{p}\neq 0} \tilde{n}(p) = n. \tag{2.27}$$

When we vary n_0, we also vary $\tilde{n}(p)$, by scaling it uniformly for all values of p, so that the total density always has the correct value.

It is known from perturbation theory[25] and from analysis of approximate ground-state wave functions[26] that for small values of p, $\tilde{n}(p)$ varies as

$$\tilde{n}(p) = n_0 M_c / 2n\hbar p. \tag{2.28}$$

for zero temperature. Here c is the velocity of sound. The numerical calculations of $\tilde{n}(p)$ by McMillan,[20] which we use, do not have this behavior, for reasons which are discussed by him and also by Chester and Reatto.[26] However, in Fig. 9 of his paper, McMillan[20] has given a prescription for modifying his numbers to agree with Eq. (2.28), and we have done that in our calculations. This particular modification has very little effect on our numerical results.

E. Relation to Other Theories

In their original paper, Cowley and Woods[5] used a theoretical model to interpret their data, and recently Puff and Tenn[6] (PT) have given a detailed theoretical analysis of the experiment done by Harling,[27] which was performed at much larger momentum and energy transfers. It is useful to compare these theories and the theory presented here.

The relation between our approach and that used by Cowley and Woods[5] has been pointed out earlier in Ref. 21, where it was noted that their model could be obtained from ours by setting the polarization potential $\psi(q)$ and the width function $\Gamma(q)$ to 0.

The experiments by Harling,[27] which Puff and Tenn[6] analyze, were done for values of q extending up to 20 Å^{-1}. These are much larger than the values of q obtained in the experiment by Cowley and Woods.[5] Puff and Tenn are also more concerned with analyzing the temperature dependence of the shape of the cross section, both above and below the superfluid transition temperature. Such an analysis cannot be carried out numerically with the theory presented here, since nothing is known about the temperature dependence of $\tilde{n}(p)$. Therefore, we restrict ourselves to a formal comparison of the two theories.

In order to relate our theory to that of Puff and Tenn it is necessary to establish the asymptotic behavior of $\psi(q)$ for large values of q. It is difficult to do this rigorously, and the following analysis can only be considered suggestive.

It seems physically reasonable that $\psi(q)$ should vanish for $q \to \infty$. This means that the response of the system to very-short wavelength disturbances is independent of the collective properties of the system.

It is evident from Eq. (2.23) that, for large q,

$$P_3(q) = \text{const}/q^2. \tag{2.29}$$

From this result and from the assumption that $\psi(q)$ approaches 0, we can conclude from Eq. (2.22) that for large q, $\Gamma(q)$ can possibly be increasing, but it must increase less rapidly than q^2. In particular, it will always increase less rapidly than ω_q. It then follows from Eq. (2.16) that the integral of $(-\hbar/\pi n)\,\chi''_{sc}(q, \omega)$ over positive ω approaches unity as q becomes large. Since $S(q)$ approaches unity as q becomes large, it seems plausible from Eq. (2.11) and from the expansion of Eq. (2.24) in powers of $\psi(q)$ that $\psi(q)$ must approach 0 at the

same rate that $[S(q)-1]$ does. Thus, starting from the assumption that $\psi(q)$ approaches 0 at large q, we have been able to make an estimate of how rapid that approach is.

Since the pair correlation function $g(r)$ is not singular at $r=0$, $[S(q)-1]$ and hence $\psi(q)$ must approach 0 more rapidly than q^{-2}. Thus we have that in the region of q values of interest to Puff and Tenn, it is a good approximation to take

$$\psi(q)=0 \quad (q \gtrsim 9\ \text{Å}^{-1}). \tag{2.30}$$

We then have from Eqs. (2.22) and (2.29) that

$$\lim_{q\to\infty}\Gamma(q)=\text{const}>0. \tag{2.31}$$

That is, $\Gamma(q)$ actually does not increase but approaches a constant value at large q, if the above argument is correct. The behavior of $\psi(q)$ and $\Gamma(q)$ given in Eqs. (2.30) and (2.31) is in agreement with our numerical results in Sec. III, but again that is only suggestive and not conclusive.

Equation (2.30) is the first step in relating our theory to Puff and Tenn. The next step is to note, following them, that for this range of q values $P_3(q)$ is quite small compared to the other terms in the third moment Eq. (2.9). Therefore, they require satisfaction of the approximate third moment

$$P_3(q)=0 \tag{2.32}$$

in Eq. (2.9). If we make this approximation in our theory, it then follows that the "constant" in Eqs. (2.29) and Eq. (2.31) is 0 and therefore

$$\Gamma(q)=0. \tag{2.33}$$

From Eqs. (2.14), (2.16), (2.30), and (2.33) it is then seen that $\chi''(q,\omega)$ has been reduced back to the free-particle form except that the exact $\tilde{n}(p)$ function appears in it. This is then essentially the form used by Cowley and Woods[5] to analyze their data at the smaller-q values.

The next assumption that must be made to obtain the Puff and Tenn result is that $\tilde{n}(p)$ has a Gaussian shape, i.e.,

$$\tilde{n}(p)=Ae^{-\alpha^2p^2}. \tag{2.34}$$

The two parameters A and α can be expressed in terms of the density of noncondensed particles $n-n_0$ and the kinetic energy per particle $\langle \text{KE}\rangle/N$. With this simple form for $\tilde{n}(p)$, the result for $\chi''(q,\omega)$ is

$$\begin{aligned}\chi''(q,\omega)=&-n_0(\pi/\hbar)[\delta(\omega-\omega_q)-\delta(\omega+\omega_q)]\\&-(n-n_0)(\pi/\hbar)[\pi\Gamma_2(q)]^{-1/2}\\&\times[e^{-(\omega-\omega_q)^2/\Gamma_2(q)}-e^{-(\omega+\omega_q)^2/\Gamma_2(q)}],\end{aligned} \tag{2.35}$$

with

$$\Gamma_2(q)=\frac{1}{1-(n_0/n)}\,\frac{8}{3\hbar}\,\frac{\langle \text{KE}\rangle}{N}\,\omega_q. \tag{2.36}$$

The expression in Eq. (2.35) satisfies the moment relations in the same approximation used by Puff and Tenn. That is, Eq. (2.8) is satisified, Eq. (2.9) is satisfied except for the absence of the last term on the right-hand side, which is small, and the value of the integral in Eq. (2.11) corresponds to $S(q)$ equal to unity, which is appropriate for the large-q values of interest to Puff and Tenn.

Equations (2.35) and (2.36) are not quite the same as the form for the cross section assumed by Puff and Tenn (PT). In order to obtain their form, we must broaden the δ functions in the first term of Eq. (2.35) into Gaussians with a width function $\Gamma_1^{\text{PT}}(q)$, so that

$$\begin{aligned}\chi''(q,\omega)=&-n_0(\pi/\hbar)[\pi\Gamma_1^{\text{PT}}(q)]^{-1/2}\\&\times\{\exp[-(\omega-\omega_q)^2/\Gamma_1^{\text{PT}}(q)]\\&-\exp[-(\omega+\omega_q)^2/\Gamma_1^{\text{PT}}(q)]\}\\&-(n-n_0)(\pi/\hbar)[\pi\,\Gamma_2^{\text{PT}}(q)]^{-1/2}\\&\times\{\exp[-(\omega-\omega_q)^2/\Gamma_2^{\text{PT}}(q)]\\&-\exp[-(\omega+\omega_q)^2/\Gamma_2^{\text{PT}}(q)]\}.\end{aligned} \tag{2.37}$$

In order for Eq. (2.37) to satisfy the same third moment relation that Eq. (2.35) satisfies, it is necessary that the width function in the second term $\Gamma_2^{\text{PT}}(q)$ be reduced below the value of $\Gamma_2(q)$ given in Eq. (2.36). The amount of reduction necessary is determined by what is chosen for $\Gamma_1^{\text{PT}}(q)$, which must be determined from a separate argument not depending on the moment relations.

Puff and Tenn choose

$$\Gamma_1^{\text{PT}}(q)=\frac{1}{2\ln 2}\,\frac{(n\sigma)^2}{M}\,\omega_q, \tag{2.38}$$

where σ is the cross section for scattering of two helium atoms. This form was suggested by Hohenberg and Platzman[4] and is derived from the finite lifetime of a single-particle state due to binary collisions with other atoms. With this expression for $\Gamma_1^{\text{PT}}(q)$, the other width function is

$$\Gamma_2^{\text{PT}}(q)=\frac{1}{1-(n_0/n)}\left(\frac{8}{3\hbar}\,\frac{\langle \text{KE}\rangle}{N}-\frac{n_0}{n}\,\frac{1}{2\ln 2}\,\frac{(n\sigma)^2}{M}\right)\omega_q. \tag{2.39}$$

Equations (2.37)–(2.39) are the ones used by Puff and Tenn.

We conclude that the form for the density response function assumed by Puff and Tenn can almost be obtained from our theory if we approximate the third moment in the same way that they do, and assume that $\tilde{n}(p)$ is a Gaussian function. The one feature of their theory which cannot be obtained from ours is the width function $\Gamma_1^{\text{PT}}(q)$ given in Eq.

(2.38), which must be introduced from other considerations.

In our theory we have not made the assumption that $P_3(q)$ is 0 and, therefore, we obtain the behavior of $\Gamma(q)$ given in Eq. (2.31) rather than that of Eq. (2.33).

In this large-q region where Eq. (2.30) holds, our result for $\chi''(q, \omega)$ in Eq. (2.24) becomes the expression in Eq. (2.25). The first term is a Gaussian with a width determined by $\Gamma(q)$. From Eq. (2.31) this width approaches a constant value at large q. From the numerical results presented in Sec. III B, it will be seen that the width of the second term in Eq. (2.25) continues to increase as q increases. Thus, at sufficiently large q our calculated cross section obtains the qualitative shape originally proposed by Hohenberg and Platzman, namely a distinct peak due to scattering from particles in the condensate sitting on top of a smooth background due to scattering from the noncondensed particles. Our calculations indicate that this situation should be obtained for $q \gtrsim 20$ Å^{-1}. In contrast, in the theory of Puff and Tenn both width functions $\Gamma_1^{PT}(q)$ and $\Gamma_2^{PT}(q)$ increases proportionally to q^2, and for that reason they do not get a separation of the cross section into two observably distinct terms.

The occurrence of this distinct separation in our theory is due to our assumption that $\psi(q)$ vanishes sufficiently rapidly. If this is not the case then $\Gamma(q)$ will increase with q and the separation will be less distinct. However, as long as $\psi(q)$ vanishes, $\Gamma(q)$ cannot increase as fast as q^2, and hence in our theory the condensate contribution will become visibly distinct at sufficiently large q. Thus, there is a definite difference between the predictions of our theory and that of Puff and Tenn concerning the visibility of the condensate peak in large momentum transfer inelastic neutron scattering. Further experiments will help resolve this difference.

Although both our theory and the theory of Puff and Tenn are phenomenological, we would like to mention four ways our theory differs from theirs. First, the resonance form of our assumed response function, Eq. (2.14), provides a mathematical mechanism for producing a shift of the maximum of the cross section away from the free-particle recoil energy, whereas their theory does not provide for such a possibility. Second, both theories take into account effects of the interparticle interactions by broadening the δ functions which are characteristic of free-particle response. However, Puff and Tenn do this by broadening only the δ functions for the particles in the condensate in Eq. (2.15) by their function $\Gamma_1^{PT}(q)$, whereas we broaden all the δ functions in Eq. (2.15) by our function $\Gamma(q)$. Physically, they take into account the effect of the interactions only on the particles which are initially in the condensate and are scattered out of it by the external disturbance, whereas we consider the effect on all the particles. The main contribution to their function $\Gamma_2^{PT}(q)$ comes from having a distribution of momenta, given by $\tilde{n}(p)$, and not directly from the effects of the interactions. Third, our theory does not make use of Eq. (2.38). In the absence of a first-principles theory, we feel that the width functions are more reliably determined using only the moment conditions, which describe exactly the short-time evolution of the system. Lastly, it was pointed out that their form for the cross section resulted from assuming a Gaussian form for $\tilde{n}(p)$. We will indicate in Sec. III A that our calculated results depend fairly sensitively on the behavior of $\tilde{n}(p)$, especially for large p, and that assuming a Gaussian form gives significantly different results than are obtained using McMillan's computed $\tilde{n}(p)$.

III. RESULTS

A. 2.5 Å^{-1} $\leq q \leq$ 9 Å^{-1}

In Sec. II it was shown that data for the static structure factor $S(q)$ and its Fourier transform $g(r)$ are needed in two different places in this calculation. The evaluation of $P_3(q)$ in Eq. (2.22) requires $g(r)$, and the satisfaction of Eq. (2.11) requires comparison with $S(q)$. These details are discussed in the Appendix. Here it suffices to say that we present two sets of calculations, one using the experimental results of Achter and Meyer (AM)[28] and the other using the theoretical results of Schiff and Verlet (SV).[23] The latter authors calculated $g(r)$ and $S(q)$ from an approximate ground-state wave function of the Jastrow form, using molecular-dynamics techniques to evaluate the required integrals. For reasons which we give in the Appendix, we feel that our calculation using the SV data is the more accurate one, but we give both sets of results to show the sensitivity of the calculations to the input values of $P_3(q)$.

Our calculated results for the width at half-maximum of $S(q, \omega)$ as a function of q and for three different values of n_0/n are shown in Fig. 1, along with the experimental results of Cowley and Woods.[5]

Our calculated results for the width do show some structure. This structure arises from the combined effects of the structure in $S(q)$ and $P_3(q)$ showing up in $S(q, \omega)$ when we require satisfaction of the moment relations. The magnitude of our calculated widths generally agrees with the data. In particular, we obtain a rapid increase in the width between $q = 2.5$ Å^{-1} and $q = 5$ Å^{-1}, a minimum at $q = 6$ Å^{-1}, and then a less rapid increase beyond that. The experimental widths have a rapid increase between $q = 2.5$ Å^{-1} and $q = 4$ Å^{-1}, a minimum at $q = 5$ Å^{-1}, another rapid increase up to 7 Å^{-1}, and then a slower

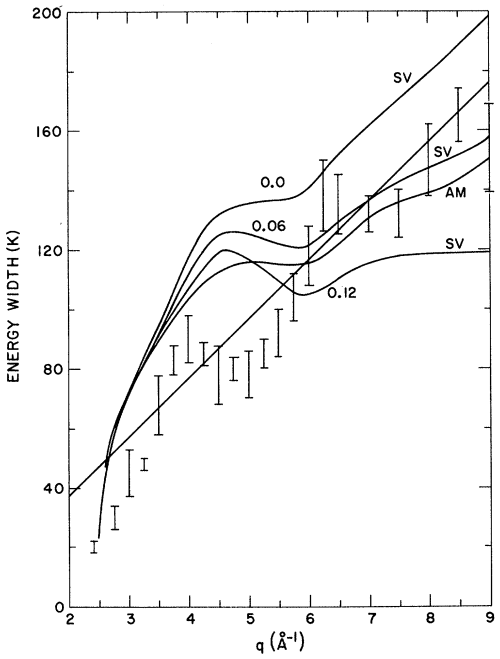

FIG. 1. Width of the cross section at half-maximum. The value used for n_0/n is marked on the curves. The experimental points are from Ref. 5. The straight line is the calculated result if $\psi(q) \equiv \Gamma(q) \equiv 0$. The curves labeled SV and AM are the calculated results using the pair distribution function of Refs. 23 and 27, respectively.

increase beyond that. It is worth mentioning that the initial rapid rise in the width is due to the presence of $\psi(q)$ in the denominator of Eq. (2.24).

The most pronounced disagreement is the presence of a definite minimum in the experimental widths at $q = 5$ Å^{-1}, where the theoretical widths have a maximum. There is a shallow minimum in the calculated widths, but it occurs at $q = 6$ Å^{-1}.

It seems reasonably conclusive from Fig. 1, especially from the larger values of q, where the theory should be most accurate, that n_0/n is greater than 0, and probably less than 0.12. Using the SV data and the procedure described in Sec. II D we obtain 0.06 as the best value of n_0/n; the AM data require a slightly lower value for the best fit.[29] These values are obtained by an "eyeball" fit to the data; the error bars are sufficiently large that a more precise fit cannot be made.

In the other calculations shown in this section we have taken $n_0/n = 0.06$. This value is essentially the same as that obtained by Puff and Tenn[6] from their analysis of the data at higher momentum transfers obtained by Harling.[27] This value is less than those obtained in theoretical calculations by McMillan,[20] by Schiff and Verlet,[23] and by Francis, Chester, and Reatto.[24] All of these calculations are based on approximate ground-state wave functions of the Jastrow type, and they obtain values of n_0/n in the range 0.08–0.11. Some of this difference may arise because we are fitting a zero-temperature theory to an experiment done at 1.1 K.

The results for the frequency of the maximum in $S(q, \omega)$, ω_M, relative to the free-particle recoil energy $\omega_q = \hbar q^2/2M$ are given in Fig. 2. This shift arises from the denominator of our expression for $S(q, \omega)$. Thus, ω_M is significantly different from ω_q when $|\psi(q)|$ is large, and ω_M/ω_q rapidly approaches unity as $|\psi(q)|$ becomes small. The three values of the experimentally measured shift reported by Cowley and Woods[5] are shown in Fig. 2, also. The agreement is fair.

Figure 3 shows the values for $\psi(q)$ obtained by satisfying Eqs. (2.9) and (2.11). For the range of q values of interest, $\psi(q)$ is negative and of such magnitude that the denominator of Eq. (2.23) shows no tendency to vanish. Thus there are no well-defined collective density oscillations in this region. Also $\psi(q)$ practically vanishes for $q \gtrsim 6.5$ Å^{-1}, which is consistent with our procedure of assuming it to be 0 when determining the value of n_0/n from the data at $q = 9$ Å^{-1}.

The width function $[\Gamma(q)]^{1/2}$ is also shown in Fig. 3. It is a rapidly varying function of q over the range where $\psi(q) \neq 0$. As was mentioned following Eq. (2.16), in a more complete theory, $\Gamma(q)$ would not be a function of q alone, but would have frequency dependence, which we have averaged out in some unknown way. The strong dependence of our $\Gamma(q)$ on q is probably a result of this averaging.

These calculations are fairly sensitive to the shape of $\tilde{n}(p)$ and thus could be modified if McMillian's[20] calculated $\tilde{n}(p)$ is not a good representation of the momentum distribution function of real liquid helium. For example, in the early stages of

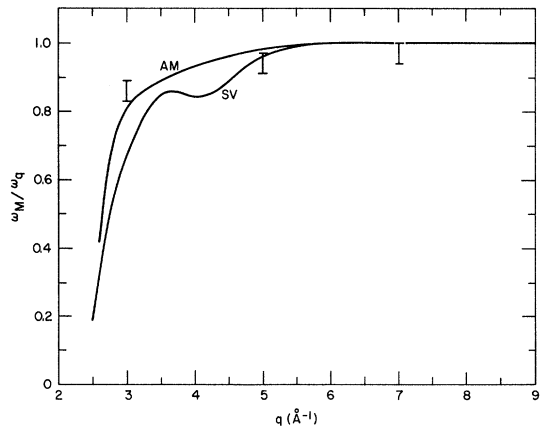

FIG. 2. Ratio of the frequency ω_M of the maximum in the cross section to the free-particle recoil $\omega_q = \hbar q^2/2M$. The experimental points are from Ref. 5 and are at $T = 1.1$ K. The calculated results are for $n_0/n = 0.06$.

this calculation we fitted McMillan's $\tilde{n}(p)$ with a Gaussian. This gives a good fit at the smaller-p values, but does not reproduce the small local maximum in McMillan's $\tilde{n}(p)$ around $p = 2.5$ Å^{-1}. According to McMillan, 15% of the atoms are in this peak. Our calculations are quite sensitive to this peak, since the quantity that enters is $p^2\tilde{n}(p)$. We found that taking this into account increased the width at the larger-q values by about 30 K.

We want to emphasize that the structure we find in the width does not depend on having a Bose-Einstein condensate, since this structure arises only from structure in the moments. Thus it should also be present if the scattering system were either liquid helium in its normal phase or a classical liquid. For the latter system this structure has been found in calculations performed by Pathak and Singwi[30] based on the classical limit of the formulas in this paper. The over-all magnitude of the width *is* affected by the presence of a condensate since the condensate tends to narrow the width at all q values as shown in Fig. 1. The amount of narrowing depends on the value of the condensate fraction n_0/n.

Since the value of the condensate fraction can be varied experimentally by changing the temperature, we expect the temperature dependence of the width to be closely related to the temperature dependence of n_0. In particular, since n_0 approaches 0 rapidly as the temperature is raised to the transition temperature, we expect to see a rapid increase in the width near the transition. This feature is present

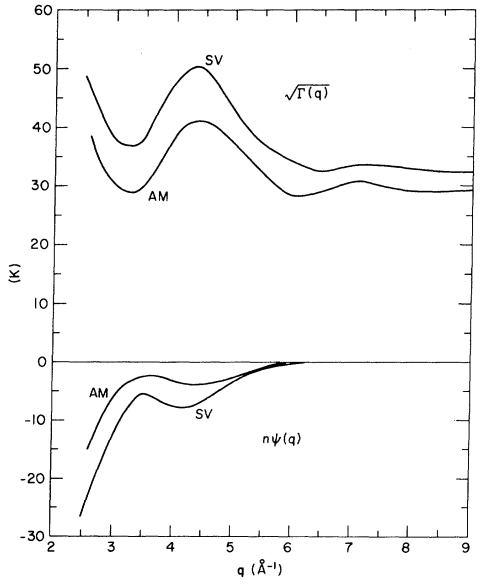

FIG. 3. Results for the width function $\Gamma(q)$ and the polarization potential $\psi(q)$ obtained by satisfying the moment relations for $n_0/n = 0.06$. The curves labeled SV and AM are the results using the pair distribution functions of Refs. 23 and 27, respectively.

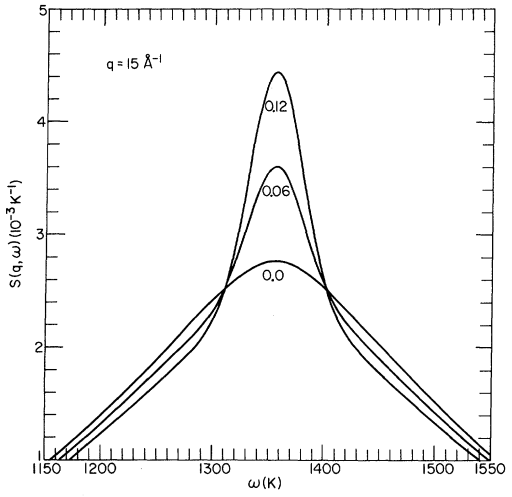

FIG. 4. Results for $S(q, \omega)$ for $q = 15$ Å^{-1} and for three different values for n_0/n, as indicated on the curves.

in the data of Cowley and Woods. To obtain quantitative results concerning this feature using our model requires a knowledge of the temperature dependence of the $\tilde{n}(p)$ function, which is not known.

B. $q \geq 9$Å^{-1}

Harling has recently published[27] some results of his neutron scattering experiments for values of q going up to 20 Å^{-1}. For that reason we give here the results of some calculations using our theory for the same range of q values. A detailed comparison with Harling's data is not given but is left for a future report, since the main purpose of this work is to compare with the results of Cowley and Woods.[5]

In Sec. II E it was shown that for sufficiently large q the $S(q, \omega)$ function given by this theory takes on the qualitative shape first proposed by Hohenberg and Platzman,[4] with a distinct peak sitting on a broad background. The numerical results presented in this section indicate how large q must be for this situation to be obtained.

In Figs. 4 and 5 we give our calculated $S(q, \omega)$ for $q = 15$ Å^{-1} and 20 Å^{-1}, respectively. For each q results are presented for three different values of n_0/n, namely 0.0, 0.06, 0.12. In Fig. 5 the dashed curve gives the contribution of just the condensate [first term in Eq. (2.25)] to $S(q, \omega)$ for $n_0/n = 0.06$.

These numerical results show that for $q = 15$ Å^{-1}, the presence of a condensate produces discernible shoulders on $S(q, \omega)$ and that for $q = 20$ Å^{-1} these shoulders are quite pronounced. This is in contrast to the situation for $q < 9$ Å^{-1}, where condensate fractions of this order of magnitude have a quantitative effect on the width of $S(q, \omega)$ but do not cause the appearance of two distinct components in

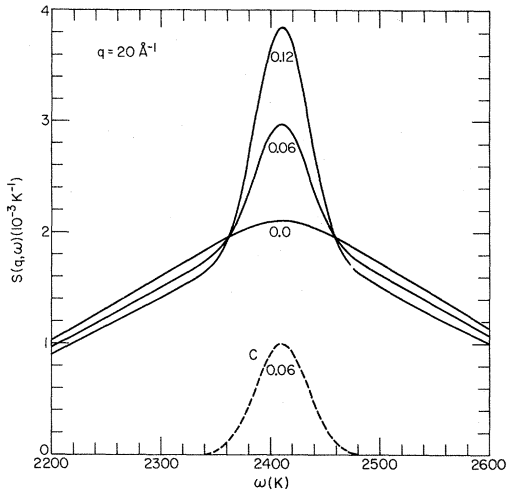

FIG. 5. Results for $S(q, \omega)$ for $q = 20$ Å^{-1} and for three different values of n_0/n, as indicated on the curves. The dashed curve is the condensate contribution only, for $n_0/n = 0.06$.

$S(q, \omega)$.

It was pointed out in Sec. II E that this situation arises because our width function $\Gamma(q)$ approaches a nonzero constant as q becomes large. Thus, the width of the condensate contribution to $S(q, \omega)$ saturates while the width of the noncondensate contribution continues to increase with increasing q.

To compare with experiment the curves in Figs. 4 and 5 must be convoluted with an appropriate resolution function. It then becomes marginal whether the shoulders can be seen at $q = 15$ Å^{-1}. However, we believe that for $q \gtrsim 20$ Å^{-1}, they will remain visible. The resolution function will have essentially no effect on the smooth background part of $S(q, \omega)$ and will broaden the condensate part slightly. Thus the shoulders should be experimentally discernible.

Although Harling[27] makes no claim in his paper to be able to see a separate contribution due to the condensate in his data, comparison of his published curve of the intensity of scattered neutrons for q around 14 Å^{-1} with our curves for $q = 15$ Å^{-1} encourages belief that the condensate contribution is probably becoming visible. In any case, experiments in this range of q values will provide a further test of this theory.

IV. CONCLUSIONS

This paper has presented a simple theory for the neutron scattering experiment at large momentum and energy transfers performed by Cowley and Woods. By imposing the conditions that the low-order moment relations be satisfied, we have been able to obtain semiquantitative agreement for the shifts and widths obtained experimentally. Although we have not conclusively demonstrated that He II does possess a Bose-Einstein condensate, we have obtained strong circumstantial evidence that it is so and have obtained a value of about 6% for the condensate fraction. We have also pointed out the interesting possibility that experiments with $q \gtrsim 20$ Å^{-1} might be able to reveal more clearly the condensate contribution to the scattered neutron intensity.

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APPENDIX

We discuss here the relative merits of the two different sets of data for $g(r)$ which we have used for calculating $P_3(q)$. The integral expression for $P_3(q)$ in Eq. (2.22) shows that the integrand is essentially $g(r)$ multiplied by derivatives of the interparticle potential $V(r)$. We take the Lennard-Jones form for $V(r)$,

$$V(r) = 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^6], \tag{A1}$$

with $\epsilon = 10.22$ K and $\sigma = 2.556$ Å. The pair correlation function is essentially zero inside the strongly repulsive part of the potential and increases rapidly outside that region. The derivatives of $V(r)$ are large in the repulsive region, and then become small at larger r. Thus the main contribution to

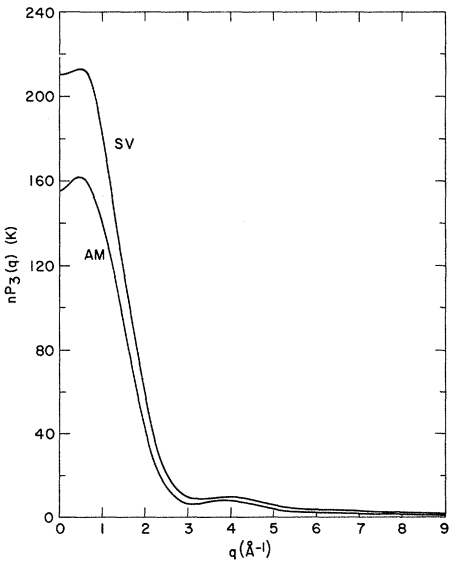

FIG. 6. Calculated values of the integral $P_3(q)$ defined in Eq. (2.23). The curves labeled SV and AM are the calculated results using the pair distribution function of Refs. 23 and 27, respectively.

the integral comes for r values close to $r=\sigma$. The value of the integral depends very sensitively on the values of $g(r)$ in this region.

It is very difficult to obtain accurate values for $g(r)$ in this region from the experimental data for $S(q)$, since it requires obtaining accurate data at large q or large scattering angles. It is just in this region that the calculations of $g(r)$ from approximate wave functions are better, because the calculations can be done in $\vec{r}$ space directly, thus avoiding the Fourier transform.

From the data for $g(r)$ given by Achter and Meyer[27] we must take $g(r)=0$ for $r<2.2$ Å. The data for $g(r)$ given by Schiff and Verlet[23] are nonzero down to $r=1.72$ Å. The results for $P_3(q)$ calculated from these two $g(r)$ functions differ by about 25%; this difference is totally due to the "core contribution" from 1.7 Å $\leq$ 2.2 Å. The two results are shown in Fig. 6.

Only the part of Fig. 6 for $q>2.5$ Å^{-1} is needed for the calculations of this paper. For these q values, the magnitude of the differences between the two sets of numbers is much smaller than at $q=0$.

For the special case of the Lennard-Jones potentail, the $q=0$ value $P_3(0)$ can be related to the average total energy per particle E/N and the average kinetic energy per particle by[31]

$$nP_3(0)=\tfrac{106}{15}\langle \mathrm{KE}\rangle/N-\tfrac{216}{15}E/N. \qquad \text{(A2)}$$

Using a calculated value[32] of $\langle \mathrm{KE}\rangle/N=14.3$ K and the experimental value[33] of $E/N=-7.14$ K gives $nP_3(0)=203.9$ K. Using Schiff and Verlet's data, we find $nP_3(0)=210.2$ K, whereas using Achter and Meyer's data we find $nP_3(0)=155.6$ K. On the basis of this comparison, we feel that our results based on Schiff and Verlet's $g(r)$ are the more reliable.

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†Present address: Department of Physics, Wake Forest University, Winston-Salem, N. C. 27109.

‡Present address: Physics Department, Punjab University, Chandigarh, Indian.

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