Momentum distributions and final-state effects in neutron scattering

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A method for analyzing neutron-scattering data at intermediate and high-momentum transfer in liquids and solids is proposed. The aim is to separate the observed dynamic structure factor, $S(Q,\omega)$, into its impulse-approximation (IA) and final-state (FS) parts. When the separation is made, both the IA and FS effects can be determined from the data. The method is based on a cumulant expansion of the intermediate scattering function, $S(Q,t)$, in powers of t. The expansion parameters are determined by fitting the corresponding $S(Q,\omega)$ to the data. Using the parameters, the IA and FS function are reconstructed. Variants of the method suitable for systems in which momentum distribution is (1) close to a Gaussian and (2) differs markedly from a Gaussian are proposed. The first variant is applied to recent data of Andersen *et al.* in normal ^{4}He at saturated vapor pressure and $T=2.5$ K and the kinetic energy, momentum distribution, and the FS broadening function are determined. In the second variant a model momentum distribution is introduced. This is applied to superfluid ^{4}He using a model $n(k)$ having a condensate. The model parameters are determined and bounds on the condensate fraction ($n_0=0.10\pm0.03$) are set. The method can be applied to any liquid, solid, or mixture. Full results for the momentum distribution and FS effects in normal and superfluid ^{4}He are presented in a forthcoming paper.

I. INTRODUCTION

The aim of neutron-scattering measurements at high-momentum transfer is to determine single-particle properties such as the momentum distribution and atomic kinetic energies in fluids and solids. Measurements at high Q were originally proposed[1,2] to observe the condensate fraction, n_0, in superfluid ^{4}He. Since that time there have been many measurements[3–20] and theoretical papers[21–30] on liquid ^{4}He and other systems. Full reference to a growing literature appears in several review articles.[31–33]

Extracting the momentum distribution from the observed coherent dynamic structure factor, $S(Q,\omega)$, however, is complicated by the interaction of the struck single particle with remainder of system; the final-state (FS) interactions. Recently, this problem has been addressed by going to higher-momentum transfer, where $S(Q,\omega)$ more nearly approaches the impulse-approximation (IA) and FS contributions are small. In this way and using calculated values of the FS broadening function[26] the condensate fraction, n_0, has been recently measured[14–18] in superfluid ^{4}He and atomic kinetic energies have been determined[15,17,20] in several quantum solids and fluids. The main features of the FS broadening function in liquid ^{4}He at high-momentum transfer ($\hbar Q$) have also been determined from experiment.[16]

In this paper, we propose a general method for analyzing neutron-scattering data at intermediate- and high-momentum transfer. The aim is to separate explicitly the IA from the FS contributions to $S(Q,\omega)$ and to determine both. The method also provides a functional form of $S(Q,\omega)$, which has a sound physical basis that can be fitted to observed data. In this way other properties of $S(Q,\omega)$ such as the peak position and full width at half maximum can be readily displayed. Rather than going to the highest reasonable Q values the method is intended for use at intermediate Q, where FS effects are not small and can therefore be identified in $S(Q,\omega)$.

The method is based on expanding the intermediate scattering function, $S(Q,t)$, in a power series in t. High Q is a short scattering time limit. Terms up to t^6 are retained in the examples presented here. The coefficients in the expansion can be related to the central moments of $S(Q,\omega)$, as discussed, for example, by Rahman, Singwi, and Sjölander[34] and Sears.[24] In the proposed method, the coefficients are treated as parameters to be determined by fitting the Fourier transform, $S(Q,\omega)$, to experiment. The IA and the FS function contribute to $S(Q,t)$ at different powers of t and with coefficients, which have a different Q dependence. The coefficients belonging to each can be identified and the $S_{\rm IA}(Q,t)$ [and therefore the one-body density matrix and $n(\mathbf{k})$] and FS function, $R(Q,\omega)$, can be constructed. For systems in which $n(\mathbf{k})$ differs markedly from a Gaussian, a model $n(\mathbf{k})$ should be introduced. In this case parameters in the model $n(\mathbf{k})$ and in $R(Q,\omega)$ are determined by fitting to experiment. The method holds in both the coherent and incoherent regimes and applies to any solid or liquid.

We begin by illustrating the procedure in the simpler incoherent limit in Sec. II. The full method for the coherent and incoherent cases is set out in Sec. III. The character of the moments of the incoherent function, $S_i(Q,\omega)$, and of $R(Q,\omega)$ are set out in Sec. IV, which tells us what form to expect for the expansion coefficients. In Sec. V, the method is illustrated for normal liquid ^{4}He, where $n(\mathbf{k})$ is expected to be approximately Gaussian. Application for a system in which $n(\mathbf{k})$ has distinct non-

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Gaussian character, superfluid ^{4}He, is discussed in Sec. VI. Full results for $S(Q,\omega)$, $n(\mathbf{k})$, and $R(Q,\omega)$ obtained using the method in normal and superfluid ^{4}He are presented in a forthcoming paper.[35]

II. ILLUSTRATION IN THE INCOHERENT LIMIT

In this section we illustrate the fitting method for the incoherent limit, where the static structure factor $S(Q)=1$ and the expansion in powers of t is simpler. We begin by expanding the impulse approximation.

The IA is defined in terms of the atomic momentum distribution $n(\mathbf{k})$ by,[36]

$$S_{\rm IA}(Q,\omega)\equiv\int d\mathbf{k}\, n(\mathbf{k})\delta\left[\omega-\omega_R-\frac{\hbar\mathbf{k}\cdot\mathbf{Q}}{M}\right] \tag{1}$$

$$=\langle\delta(\omega-\omega_R-qk_Q)\rangle\ , \tag{2}$$

where $\omega_R=\hbar Q^2/2M$ is the free-atom recoil frequency, $q=\hbar Q/M$ is the free-atom recoil velocity and $\hbar k_Q$ is the atomic momentum variable projected along Q, $k_Q=(\mathbf{k}\cdot\mathbf{Q})/Q$. The expectation value in (2) is the usual thermal average, but, since only the momentum appears, it reduces to an average over $n(\mathbf{k})$. From (2), the intermediate function in the IA is

$$\begin{aligned}S_{\rm IA}(Q,t)&=\int_{-\infty}^{\infty}d\omega\, e^{i\omega t}S_{\rm IA}(Q,\omega)\\&=\left\langle\int_{-\infty}^{\infty}d\omega\, e^{i\omega t}\delta(\omega-\omega_R-qk_Q)\right\rangle\\&=e^{-i\omega_R t}\langle e^{-iqk_Q t}\rangle=e^{-i\omega_R t}\langle e^{-ik_Q s}\rangle\ ,\end{aligned} \tag{3}$$

where $s=(\hbar Q/M)t$. $S(Q,t)$ may be viewed as the evolution of the scattering event in the fluid beginning at $t=0$. The $k_Q=k_Q(0)$ is the initial momentum of the struck atom. The neutron transfers a velocity $\hbar Q/M$ to the atom, which we use as a reduced wave vector, $q\equiv\hbar Q/M$. We also use q as the unit to go between "scaled" and unscaled variables. The "y" scaling variable is, for example,

$$y=(M/\hbar Q)(\omega-\omega_R)=(\omega-\omega_R)/q\ .$$

Assuming the struck atom is free and $\hbar Q/M\gg v_Q=\hbar k_Q/M$, the struck atom travels a distance $s=(\hbar Q/M)t=qt$ after time t. The s is the scaled length conjugate to y.

To express $S_{\rm IA}(Q,t)$ as a power series in t, we expand (3) in cumulants,[37] [see Eq. (A12)]

$$S_{\rm IA}(Q,t)=\exp(-i\omega_R t)\times\exp\left[-\frac{1}{2}\alpha_2 t^2+\frac{1}{4!}\alpha_4 t^4-\frac{1}{6!}\alpha_6 t^6+\cdots\right]\ , \tag{4}$$

where $\alpha_n=q^n\bar{\alpha}_n$ are cumulants of k_Q,

$$\begin{aligned}\alpha_2&=q^2\bar{\alpha}_2=q^2\langle k_Q^2\rangle\ ,\\\alpha_4&=q^4\bar{\alpha}_4=q^4[\langle k_Q^4\rangle-3\langle k_Q^2\rangle^2]\ ,\\\alpha_6&=q^6\bar{\alpha}_6=q^6[\langle k_Q^6\rangle-15\langle k_Q^4\rangle\langle k_Q^2\rangle+30\langle k_Q^2\rangle^3]\ .\end{aligned} \tag{5}$$

The odd cumulants vanish because $\langle k_Q^n\rangle=0$ for odd n. The even ones can be readily related to central moments of the IA,

$$I_n=\int d\omega(\omega-\omega_R)^n S_{\rm IA}(Q,\omega)=q^n\langle k_Q^n\rangle\ . \tag{6}$$

Equation (4) is the power series in t desired. $S_{\rm IA}(Q,t)$ may be viewed as a Gaussian (term in t^2) plus corrections to a Gaussian (terms in $t^4,t^6,\ldots$). Using the Gaussian term we may introduce a scattering time $\tau^2=2/\alpha_2=2/q^2\langle k_Q^2\rangle$, which is defined as the time over which $S_{\rm IA}(Q,t)$ decays to zero. During the scattering time, the struck atom travels a distance $s_\tau=q\tau=\sqrt{2}/\langle k_Q^2\rangle^{1/2}$, which is short (i.e., $\langle k_Q^2\rangle^{1/2}\sim 1.0$ Å^{-1} in liquid ^{4}He) and independent of Q. Using s_τ and (3) we see that the convergence of (4) is independent of Q and depends only on the cumulants of k_Q. For a Gaussian, $\alpha_n=0$ for $n>2$. Thus (4) provides an approximate description of $S_{\rm IA}(Q,t)$ and $n(\mathbf{k})$.

We define the final-state broadening function $R(Q,t)$ by

$$S(Q,t)\equiv S_{\rm IA}(Q,t)R(Q,t)\ . \tag{7}$$

This is the definition introduced by Gersch and Rodriguez.[22] $R(Q,t)$ is the difference between the observed coherent $S(Q,t)$ and the IA in a simple product form. This leads to a convolution for $S(Q,\omega)$,

$$S(Q,\omega)=\int d\omega' S_{\rm IA}(Q,\omega')R(Q,\omega-\omega')\ , \tag{8}$$

where

$$R(Q,\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}dt\, e^{i\omega t}R(Q,t)\ . \tag{9}$$

If the IA held exactly, clearly $R(Q,t)=1$ and $R(Q,\omega)=\delta(\omega)$.

To illustrate the method in the incoherent limit, we follow Rahman, Singwi, and Sjölander[34] and Sears[24] and write $S_i(Q,t)$ as (see Appendix A),

$$S_i(Q,t)\equiv\langle e^{-iQ\cdot r(t)}e^{iQ\cdot r(o)}\rangle \tag{10}$$

$$=\exp(-i\omega_R t)\left\langle T_t\exp\left[-iq\int_o^t dt' k_Q(t')\right]\right\rangle\ . \tag{11}$$

In (11) T_t is the time ordering operator and the expectation value is the usual thermal average. As in the IA, we expand $S_i(Q,t)$ as,

$$S_i(Q,t)=\exp(-i\omega_R t)\exp\left[-\frac{1}{2}\mu_{2i}t^2+\frac{i}{3!}\mu_{3i}t^3+\cdots-\frac{1}{6!}\mu_{6i}t^6+\cdots\right]. \tag{12}$$

To obtain this power series in t, we use a cumulant expansion of the expectation value in (11) [see Eq. (A15)] and expand $k_Q(t)$ as $k_Q(t)=k_Q+k_Q t+\cdots$ in each cumulant. The coefficients μ_{ni} can be readily determined in this way. However, it is easier to obtain the coefficients by noting that the μ_{ni} are related to the central moments of $S_i(Q,t)$,

$$M_{ni}=\int d\omega(\omega-\omega_R)^n S_i(Q,\omega)$$
$$=i^n\lim_{t\to 0}\frac{d^n}{dt^n}[e^{i\omega_R t}S_i(Q,t)], \tag{13}$$

by

$$\mu_2=M_2,\quad \mu_3=M_3,\quad \mu_4=M_4-3M_2^2,\quad \mu_5=M_5-10M_3M_2,\quad \mu_6=M_6-15M_4M_2-10M_3^2+30M_2^3. \tag{14}$$

The general structural relationship (14) between the μ_n and the moments M_n can be demonstrated by substituting (12) into (13) and carrying out the differentiation indicated. The subscript has been dropped in (14) because (14) holds for any function $S(Q,t)$ expanded as in (12). Indeed, (13) provides a method for generating the usual expression[37] for the cumulants in terms of the moments. Equation (4) is a special case of (12) for $k_Q(t')\approx k_Q(0)$ at all t'.

Using the definition (7) and comparing the expansions of $S_{\rm IA}(Q,t)$ and $S_i(Q,t)$ in (4) and (12), respectively, we obtain $R(Q,t)$ in the incoherent limit as

$$R_i(Q,t)=\exp\left[\frac{i}{3!}\beta_{3i}t^3+\frac{1}{4!}\beta_{4i}t^4-\frac{i}{5!}\beta_{5i}t^5-\frac{1}{6!}\beta_{6i}t^6+\cdots\right] \tag{15}$$

in which the coefficients are $\beta_{ni}=\mu_{ni}$ (n odd) and

$$\beta_{ni}=\mu_{ni}-\alpha_n \quad (n \text{ even}). \tag{16}$$

An important feature is that t appears in an exponential in (15) so that $R(Q,t)$ can be Fourier transformed to obtain $R(Q,\omega)$. This $R(Q,\omega)$ can then be used in the convolution form (8).

The method consists of fitting $S_i(Q,t)$ given by (12) to experiment at several Q values with the $\mu_{ni}(Q)$ regarded as adjustable parameters. This yields empirical values of the μ_{ni}, and the aim is to determine α_n and β_n separately. This can be done because (1) $\beta_{2i}=0$ so that $\mu_{2i}=\alpha_2$, (2) the $\alpha_n=0$ for odd n so that $\beta_{ni}=\mu_{ni}$ for odd n, and (3) for n even ($n\geq 4$), $\mu_{ni}=\alpha_n+\beta_{ni}$, and the Q dependence of α_n and β_{ni} differ. For example, the $\bar{\alpha}_n$ in (5) are independent of Q, while $\bar{\beta}_n=\beta_{ni}/q^n$ are proportional to Q^{-2} (n even). By plotting $\mu_{ni}(Q)$ versus Q we can determine the contributions from α_n and β_{ni}. In the examples that follow, we determined μ_n up to μ_5. Using the α_n, $S_{\rm IA}(Q,t)$ and $n(\mathbf{k})$ is constructed. The β_n are used to determine $R(Q,t)$.

This procedure can be applied directly to any system in which $n(\mathbf{k})$ is approximately Gaussian. For systems such as superfluid ^{4}He in which the IA is not well described by (4) (i.e., the one-body density matrix has a long time tail) it is important to introduce a specific model of $n(\mathbf{k})$ as discussed in Sec. V below. The method may be regarded as providing a functional form of $S(Q,\omega)$ to fit to data and a means of disentangling $R(Q,t)$ from $S_{\rm IA}(Q,t)$.

III. COHERENT CASE

In this section, we derive the method for the coherent dynamic structure factor, $S(Q,\omega)$. It is expected to be useful at Q values high enough that all collective response of the fluid or solid has disappeared but the incoherent limit has not been reached. In liquid ^{4}He this is the range $5\lesssim Q\lesssim 10$ Å^{-1}.

The coherent intermediate scattering function is[36]

$$S(Q,t)\equiv\frac{1}{N}\sum_{i,j}\langle e^{-i\mathbf{Q}\cdot\mathbf{r}_i(t)}e^{i\mathbf{Q}\cdot\mathbf{r}_j(0)}\rangle. \tag{17}$$

In Appendix A, we show this may be rearranged as

$$S(Q,t)=S(Q)\exp(-i\omega_R t)$$
$$\times\left\langle\exp\left[-iQ\int_0^t dt' v_{iQ}(t')\right]\right\rangle_S, \tag{18}$$

where the expectation value $\langle 0\rangle_S$ is defined as

$$\langle 0\rangle_S=\langle 0\hat{S}(Q)\rangle/\langle\hat{S}(Q)\rangle. \tag{19}$$

Here

$$\hat{S}(Q)=\sum_j e^{-i\mathbf{Q}\cdot(\mathbf{r}_i-\mathbf{r}_j)} \tag{20}$$

is the static structure factor operator, $\langle\ \rangle$ is the usual thermal expectation value, and $S(Q)=\langle\hat{S}(Q)\rangle$ is the usual static structure factor. In (18), v_{iQ} is the velocity of atom i along Q and, since all atoms are identical, we drop the subscript i. Clearly (18) reduces to the incoherent result (11) at high enough Q that $S(Q)\to 1$.

The expectation value in (18) is a normalized expectation value suitable for a cumulant expansion. Making this expansion [see Appendix A, (A15)], we have

$$S(Q,t)=S(Q)\exp(-i\omega_R t)\exp\left[\sum_{n=1}^{\infty}\frac{1}{n!}\mu_n(-it)^n\right]$$
$$=S(Q)\exp(-i\omega_R' t)$$
$$\times\exp\left[-\frac{1}{2}\mu_2 t^2+\cdots-\frac{1}{6!}\mu_6 t^6+\cdots\right], \quad (21)$$

where $\omega_R'\equiv\omega_R+\mu_1$. It is convenient to introduce the shifted recoil frequency $\omega_R'=\omega_R+\mu_1$. The first central moment of $S(Q,\omega)$ centered at ω_R' vanishes. To see this, note that the usual first coherent central moment is

$$\langle(\omega-\omega_R)\rangle=\int d\omega(\omega-\omega_R)S(Q,\omega)=\omega_R[1-S(Q)], \quad (22)$$

where we have used the f-sum rule, $\langle\omega\rangle=\omega_R$ and $\langle\omega_R\rangle=\omega_R S(Q)$. Substituting (21) into (13), we may show from the first derivative of $S(Q,t)$ that,

$$\frac{1}{S(Q)}\langle(\omega-\omega_R)\rangle=\mu_1\equiv\omega_R'-\omega_R, \quad (23)$$

so that $\omega_R'=\omega_R/S(Q)$. The normalized, central moments centered about ω_R' are

$$M_n\equiv\int d\omega(\omega-\omega_R')^n S(Q,\omega)/S(Q)$$
$$=i^n\lim_{t\to0}\frac{d^n}{dt^n}[e^{i\omega_R' t}S(Q,t)]/S(Q). \quad (24)$$

By substituting (21) into (24) and carrying out the differentiation indicated in (24), we may show that $M_1=0$ and the μ_n are related to the higher moments M_n as in (14). Expressions for the moments may be obtained by substituting (18) into (24). The first four moments are

$$M_0=1,$$
$$M_1=0,$$
$$M_2=Q^2\langle v_Q^2\hat{S}\rangle/S, \quad (25)$$
$$M_3=[Q^3\langle v_Q^3\hat{S}\rangle+iQ^2\langle\dot{v}_Q v_Q\hat{S}\rangle]/S,$$
$$M_4=\left[Q^4\langle v_Q^4\hat{S}\rangle+\frac{Q^2}{3M^2}\langle(\nabla v)^2\hat{S}\rangle\right]/S,$$

where $S\equiv S(Q)$. Although written in a somewhat different form, these moments are the same as those given by Rahman, Singwi, and Sjölander.[34] These moments oscillate with Q and in general are complicated. They are discussed recently for liquid ^{4}He by Stringari.[38]

Using the definition (7), and the expansions (4) and (21) for $S_{\rm IA}(Q,t)$ and $S(Q,t)$, respectively, the FS resolution function $R(Q,t)$ in the coherent regime is,

$$R(Q,t)\equiv S(Q,t)/S_{\rm IA}(Q,t)$$
$$=S(Q)\exp(-i\beta_1 t)$$
$$\times\exp\left[-\frac{1}{2}\beta_2 t^2+\frac{i}{3!}\beta_3 t^3+\cdots-\frac{1}{6}\beta_4 t^6\right], \quad (26)$$

where $\mu_1=[\omega_R/S(Q)][1-S(Q)]$ and

$$\beta_n=\mu_n-\alpha_n. \quad (27)$$

The μ_n and α_n are defined in (14) and (5), respectively. In this way the FS resolution function is completely specified. In the incoherent regime we expect the $\beta_n(Q)$ to oscillate with Q. These oscillations will disappear at higher Q when the incoherent limit is reached. In this limit, the central moments simplify and the Q dependence of them can be specified. Since the Q dependence of the α_n is known, the Q dependence of the β_n can then be obtained from (27) in this limit. This is done in the next section.

We conclude by setting out the moments of $R(Q,t)$. Equation (14) is a very general relation between the central moments M_n of the type defined in (24), and the coefficients μ_n of the same function expanded in the form (21). Applying this to $R(Q,t)$ in (26), the moments are defined as

$$R_n=\frac{1}{S(Q)}\int d\omega[\omega-\mu_1]^n R(Q,\omega) \quad (28)$$

and the R_n may be readily obtained by inverting (14), i.e., $R_0=1$, $R_1=0$, and,

$$R_2=\beta_2,$$
$$R_3=\beta_3,$$
$$R_4=\beta_4+3\beta_2^2, \quad (29)$$
$$R_5=\beta_5+10\beta_3\beta_2,$$
$$R_6=\beta_6+15\beta_4\beta_2+10\beta_3^2+15\beta_2^3.$$

In the incoherent limit, where $S(Q)=1$, $\mu_1=0$, and $M_2=I_2$, we have $R_2=0$ and simplified higher moments given by (39) below.

IV. INCOHERENT MOMENTS AND $R(Q,t)$

The central moments M_{ni} of $S_i(Q,\omega)$ are defined in (13). Using these moments, the aim is to obtain simple results for the expansion coefficients μ_{ni} in (12) and the β_{ni} in (15), particularly their Q dependence.

The M_{ni} can be calculated straightforwardly by expanding the exponential in (11) [see (A13)] and carrying out the differentiation in (13). Up to M_{6i} we obtain

$$M_{ni}=I_n+A_n+B_n \quad (n\le6). \quad (30)$$

Here $I_n=q^n\langle k_Q^n\rangle$ are the moments (6). The A_n originate from the term proportional to Q^2 in the expansion of (11) [see (A14)] and $A_n=0$ for $n\le2$. The B_n originate from the term proportional to Q^4 in (11), and $B_n=0$ for $n\le4$. Clearly $M_{0i}=1$, $M_{1i}=0$, and $M_{2i}=I_2$ are the same as in the IA (6). Differentiating α_2 in (A14), we obtain

$$A_n=-i^n q^2\lim_{\omega\to0}\left\langle\frac{d^{n-2}}{dt^{n-2}}k_Q(t)k_Q(0)\right\rangle, \quad (31)$$

of which the lowest two are

$$A_3 = iq^2\langle \dot{k}_Q k_Q \rangle = q^2\langle \nabla^2 v(r)\rangle/6\hbar \equiv q^2\bar{a}_3 ,$$
$$A_4 = -q^2\langle \ddot{k}_Q k_Q \rangle = q^2\langle F_Q^2\rangle/3\hbar^2 \equiv q^2\bar{a}_4 , \tag{32}$$

where $F_Q = \nabla_Q v(r)$ is the force on the struck atom along the scattering vector Q. The higher terms are more complicated and are written in Appendix B. To display the Q dependence, we write them as

$$A_5 = q^2\bar{a}_{52}, \quad B_5 = q^4\bar{b}_{54} ,$$
$$A_6 = q^2\bar{a}_{62}, \quad B_6 = q^4\bar{b}_{64} . \tag{33}$$

The coefficients μ_n can be obtained from the moments M_n using the general relation (14). Using (14) to relate μ_{ni} to M_{ni} and α_n to I_n, we have

$$\mu_{2i} = q^2\bar{\alpha}_2 = q^2\langle k_Q^2\rangle ,$$
$$\mu_{3i} = q^2\bar{a}_3 ,$$
$$\mu_{4i} = q^2\bar{a}_4 + q^4\bar{\alpha}_4 , \tag{34}$$
$$\mu_{5i} = q^2\bar{a}_{52} + q^4\bar{a}_{54} ,$$
$$\mu_{6i} = q^2\bar{a}_{62} + q^4\bar{a}_{64} + q^6\bar{\alpha}_6 ,$$

where the $\bar{\alpha}_n$ are defined in (5), and

$$\bar{a}_3 = \langle \nabla^2 v(r)\rangle/6\hbar ,$$
$$\bar{a}_4 = \langle F_Q^2\rangle/3\hbar^2 ,$$
$$\bar{a}_{54} = \bar{b}_{54} - 10\bar{a}_3\langle k_Q^2\rangle , \tag{35}$$
$$\bar{a}_{64} = \bar{b}_{64} - 15\bar{a}_4\langle k_Q^2\rangle - 10\bar{a}_3^2$$

are independent of q. The coefficients $\beta_{ni} = \mu_{ni} - \alpha_n$ are (34) without the $\alpha_n = q^n\bar{\alpha}_n$.

The $\bar{\alpha}_2 = \langle k_Q^2\rangle$ must be positive, but the $\bar{\alpha}_4$ and $\bar{\alpha}_6$ can be positive or negative. For a solid in which the dynamics can be represented by a density of phonon states $g(\omega)$ (31) can be readily expressed in terms of the moments of $g(\omega)$,

$$A_n = \omega_R\langle \omega^{n-1}\rangle , \tag{36}$$

where

$$\langle \omega^n\rangle = \begin{cases} \int d\omega\, g(\omega)\omega^n[2n_B(\omega)+1], & n-\text{odd} , \\ \int d\omega\, g(\omega)\omega^n, & n-\text{even} , \end{cases} \tag{37}$$

and $n_B(\omega)$ is the Bose function. The moments (37) must be positive so that the $\bar{a}_n$ are positive. In a fluid $\langle \nabla^2 v(r)\rangle/M$ represents an average frequency, which is positive, and $\langle F_Q^2\rangle$ is positive. A positive $\mu_{3i} = M_{3i}$ shifts the peak of $S_i(Q,\omega)$ to lower ω (below ω_R), which appears to be universally observed. For a classical fluid in which $n(\mathbf{k})$ is Gaussian (Maxwell-Boltzmann distribution) and in a solid in which the phonons are statistically independent and a $g(\omega)$ can be defined, we find (see also Appendix B and Sears[21]),

$$\bar{b}_{54} = 10\bar{a}_3\langle k_Q^2\rangle ,$$
$$\bar{b}_{64} = 15\bar{a}_4\langle k_Q^2\rangle + 10\bar{a}_3^2 . \tag{38}$$

In this case, $\bar{a}_{54} = \bar{a}_{64} = 0$, and the terms in μ_{5i} and μ_{6i} proportional to q^4 vanish. The only final-state terms remaining in μ_{ni} are the A_n terms. We have, however, found that in normal and superfluid ^{4}He that the term in μ_{5i} proportional to Q^4 dominates so that this is apparently not a good approximation (i.e., $n(\mathbf{k})$ is not a Gaussian in a quantum fluid).

The moments of $R(Q,\omega)$ in (28) from the incoherent limit, where $S(Q)=1$ and $\mu_1 = 0$ are $R_{0i} = 1$, $R_{1i} = R_{2i} = 0$, and

$$R_{3i} = M_{3i} = q^2\langle \nabla^2 v(r)\rangle/6\hbar ,$$
$$R_{4i} = A_4 = q^2\langle F_Q^2\rangle/3\hbar^2 ,$$
$$R_{5i} = M_{5i} - 10A_3I_2 = A_5 + B_5 - 10A_3I_2 , \tag{39}$$
$$R_{6i} = A_6 + B_6 - 15A_4I_2 .$$

Using these moments, we may write the μ_{ni} as

$$\mu_{2i} = I_2 ,$$
$$\mu_{3i} = R_{3i} ,$$
$$\mu_{4i} = R_{4i} + (I_4 - 3I_2^2) , \tag{40}$$
$$\mu_{5i} = R_{5i} ,$$
$$\mu_{6i} = R_{6i} - 10R_{3i}^2 + (I_6 - 15I_4I_2 + 30I_2^3) ,$$

which displays their dependence on the moments of the IA and on FS effects.

V. APPLICATION FOR NEARLY GAUSSIAN $n(\mathbf{k})$: NORMAL LIQUID ^{4}He

The fitting procedure outlined above in Sec. III can be applied directly to any fluid or solid in which the momentum distribution is approximately Gaussian. The $S(Q,t)$ given by (21), i.e.,

$$S(Q,t) = S(Q)\exp(-i\omega_R' t)\exp\left[-\frac{1}{2}\mu_2 t^2 + \frac{i}{3!}\mu_3 t^3 + \frac{1}{4!}\mu_4 t^4 - \frac{i}{5!}\mu_5 t^5 - \frac{1}{6!}\mu_6 t^6\right] , \tag{41}$$

is fitted to experiment to obtain the coefficients μ_n and the FS resolution function $R(Q,t)$ is constructed from (26) and (27). $S(Q,\omega)$ and $R(Q,\omega)$ are obtained by Fourier transform. In the incoherent limit, $S(Q)=1$ and $\omega_R' = \omega_R/S(Q) \to \omega_R$. The expansion (41) and the expansion of $S_{\mathrm{IA}}(Q,t)$ in (4) introduce a model for $n(\mathbf{k})$. The model is a Gaussian, from the t^2 term in (4) plus corrections from higher powers in t. We apply the method to normal liquid ^{4}He ($T > T_\lambda$), where we expect $n(\mathbf{k})$ can be represented by a Gaussian plus corrections to the Gauss-

ian. We use two variants of the method beginning with (41). In the first, the terms in the second exponent beyond the t^2 in (41) are expanded. The $S(Q,t)$ is then a Gaussian plus additions, denoted the additive approach (AA). The second variant is a straightforward application of the full function (41) and is denoted the convolution approach (CA).

A. Additive approach

In the additive approach, the general expression (41) for $S(Q,t)$ to be fitted to experiment is expanded as

$$S(Q,t)\simeq S(Q)e^{-i\omega_R' t}e^{-(1/2)\mu_2 t^2} \times\left[1+\frac{i}{3!}\mu_3 t^3+\frac{1}{4!}\mu_4 t^4-\frac{i}{5!}\mu_5 t^5-\cdots\right]. \quad (42)$$

In (42), we have expanded the cubic and higher-power terms in t and retained the terms up to $O(t^5)$. The corresponding expansion for $S_{\rm IA}(Q,t)$ in (4) is

$$S_{\rm IA}(Q,t)=\exp(-i\omega_R t)\exp\left[-\frac{1}{2}\alpha_2 t^2+\frac{1}{4!}\alpha_4 t^4-\cdots\right] \simeq e^{-i\omega_R t}e^{-(1/2)\alpha_2 t^2}\left[1+\frac{1}{4!}\alpha_4 t^4-\cdots\right]. \quad (43)$$

The Fourier transform,

$$S(Q,\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}dt\, e^{i\omega t}S(Q,t) \quad (44)$$

of (42) takes a simple, additive analytic form,

$$S(Q,\omega)=\hat{S}_{\rm IA}(Q,\omega)+S_1(Q,\omega)+S_2(Q,\omega)+S_3(Q,\omega)\ , \quad (45)$$

where

$$\hat{S}_{\rm IA}(Q,\omega)=S(Q)[2\pi\mu_2]^{-1/2}e^{-(\omega-\omega_R')^2/2\mu_2}\ ,$$
$$S_1(Q,\omega)=-\frac{\mu_3}{2\mu_2^2}(\omega-\omega_R')\left[1-\frac{\omega_d^2}{3}\right]\hat{S}_{\rm IA}(Q,\omega)\ ,$$
$$S_2(Q,\omega)=\frac{\mu_4}{8\mu_2^2}\left[1-2\omega_d^2+\frac{\omega_d^4}{3}\right]\hat{S}_{\rm IA}(Q,\omega)\ , \quad (46)$$
$$S_5(Q,\omega)=\frac{\mu_5}{8\mu_2^3}(\omega-\omega_R')\left[1-\frac{2\omega_d^2}{3}+\frac{\omega_d^4}{15}\right]\hat{S}_{\rm IA}(Q,\omega)\ ,$$

and $\omega_R'=\omega_R/S(Q)$ and $\omega_d^2=(\omega-\omega_R')^2/\mu_2$.

We recall that in the incoherent limit, the coefficients μ_n in (46) are ($\mu_n\rightarrow\mu_{ni}$) given by (34). In the incoherent limit, $\hat{S}_{\rm IA}(Q,\omega)$, is a Gaussian impulse approximation with $\mu_2=\alpha_2$. The $S_1(Q,\omega)$ and $S_3(Q,\omega)$ represent deviations due solely to final-state contributions, $\mu_3=\beta_3$, $\mu_5=\beta_5$.

The $S_2(Q,\omega)$, with $\mu_4=\beta_4+\alpha_4$, represents a deviation of $n(\mathbf{k})$ from a Gaussian, due to α_4, plus final-state effects. Clearly, $S_1(Q,\omega)$ and $S_3(Q,\omega)$ are odd in $(\omega-\omega_R')$ and $S_2(Q,\omega)$ is even in $(\omega-\omega_R)$. In the incoherent limit, the expansion (45) is the same as proposed by Sears[24] except that (45) uses a Gaussian IA rather than the exact IA. An expansion about the exact IA contains only corrections due to FS terms.

In the additive approach, we fit (45) directly to the data with the aim of determining μ_2–μ_5. The advantage of (45) is that the largest term, $\hat{S}_{\rm IA}(Q,\omega)$, is a Gaussian, and the corrections to this Gaussian appear as simple analytic expressions. The parameters μ_2, μ_3, μ_4, and μ_5 are adjusted to get the best fit. $S(Q)$ and $\omega_R'=\omega_R/S(Q)$ are known. The disadvantage of (45) is that we cannot recover $R(Q,t)$ in a form suitable for a convolution. As usual, $S(Q,\omega)$ must be convoluted with the instrument resolution function $I(\omega)$,

$$O(Q,\omega)=\int d\omega' S(Q,\omega')I(\omega-\omega')\ , \quad (47)$$

and $O(Q,\omega)$ fitted to the observed data. In these fits, we have used both a numerically computed instrument resolution and a Gaussian approximation, $I(t)=\exp(-t^2\sigma_I^2/2)$.

A fit of the AA, to data of Andersen[39] and Andersen *et al.*[40] at $Q=8$ Å^{-1} in normal ^{4}He at $T=2.5$ K is shown in Fig. 1. The data was obtained using the MARI instrument at ISIS, Rutherford Appleton Laboratory. In the fit, only μ_2, μ_3, and μ_4 were retained because includ-

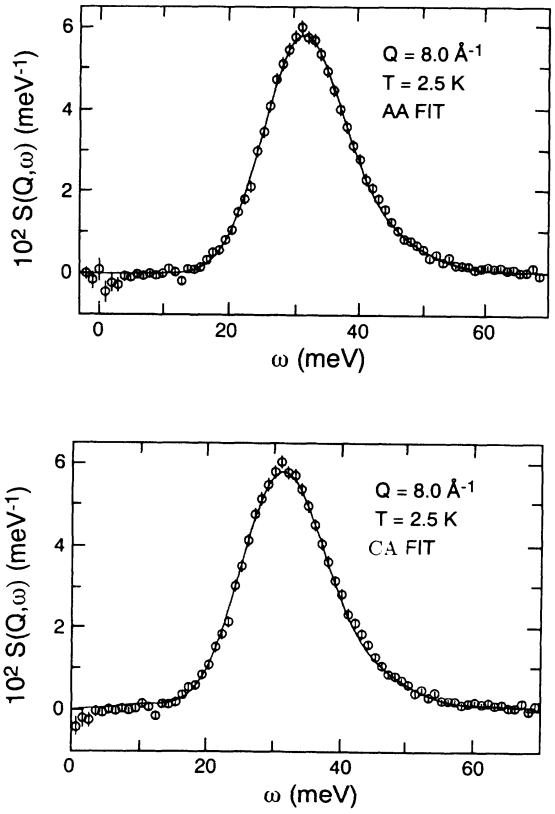

FIG. 1. Upper: Additive approach (AA), (45) and (46), fitted to data of Andersen *et al.* (Ref. 40) at $Q=8$ Å^{-1} and $T=2.5$ K. Lower: Convolution approach (CA) (41), fitted to the same data. The values of the fitted parameters are listed in Table I. A numerical instrument resolution function computed by Andersen *et al.* (Ref. 40) was used in (47). The χ^2 of the fit were (AA) $\chi^2=0.88$ and (CA) $\chi^2=0.96$ (see Appendix D).

ing μ_5 did not improve the fit (see Appendix D). Clearly, it is possible to obtain a good fit. The parameters μ_2, μ_3, and μ_4 obtained in the AA fit at $Q=8$ Å^{-1} and $Q=14$ Å^{-1} are listed in Table I.

Using data at low Q, $5 \lesssim Q \lesssim 10$ Å^{-1}, Andersen *et al.*[41] fitted the AA to many Q values. These fits led to values of $\mu_2(Q)$, $\mu_3(Q)$, and $\mu_4(Q)$ that oscillated with Q. Andersen *et al.*[41] also displayed their results as oscillations in the peak position and width of $S(Q,\omega)$ along the lines discussed previously by Martel *et al.*[9] and Stirling *et al.*[41,42] For $Q \gtrsim 10$–12 Å^{-1}, the oscillations cease and the incoherent regime is apparently reached. For $Q \gtrsim 10$–12 Å^{-1}, $\mu_2(Q)/Q^2$, $\mu_3(Q)/Q^2$, and $\mu_4(Q)/Q^4$ reached constant values.[41] From (34) (the incoherent limit) $\mu_{2i}=\alpha_2=q^2\langle k_Q^2\rangle$, which gives α_2 directly. With $\alpha_3=0$, $\beta_3=\mu_{3i}$. From (34), $\mu_{4i}=\beta_{4i}+\alpha_4$ with α_4 proportional to Q^4 and $\beta_{4i} \propto Q^2$. A plot of $\mu_4(Q)/Q^4$ reaches a constant for $Q \gtrsim 10$ Å^{-1} and shows that μ_4 is dominated by α_4 with no apparent contribution from β_{4i} (Refs. 40 and 41). In this way α_2 and α_4 were determined from the high Q values of $\mu_{2i}(Q)$ and $\mu_{4i}(Q)$ and are listed in Table II. Throughout, we use μ_n in energy units (meV). The corresponding mean-square momentum along Q is then $k_0^2 \equiv \langle k_Q^2\rangle_N=(\hbar^2/M)^{-2}(\alpha_2/Q^2)$, the kinetic energy $\langle K\rangle_N=(3/2)(\hbar^2/M)^{-1}(\alpha_2/Q^2)$ and the excess $\delta=\alpha_4/\alpha_2^2$ ($\hbar^2/M=1.0443$ meV Å^2 for ^{4}He).

B. Momentum distribution

One of the goals of the fitting procedure is to determine the momentum distribution, $n(\mathbf{k})$. The factor $\langle e^{-ik_Q s}\rangle$ in the IA in (3) is the one-body density matrix (OBDM) for displacements $s=(\hbar Q/M)t=qt$ of the struck atom along Q, $n^{-1}\rho_1(s\hat{\mathbf{Q}},0)=\langle e^{-ik_Q s}\rangle$, where $\hat{\mathbf{Q}}$ is a unit vector. The momentum distribution $n(\mathbf{k})$ is the Fourier transform of the OBDM, $n^{-1}\rho_1(\mathbf{r},0)=\langle e^{-i\mathbf{k}\cdot\mathbf{r}}\rangle$. Specifically, the longitudinal momentum distribution $n(k_Q)$, $n(\mathbf{k})$ projected along Q, is the Fourier transform of $\langle e^{-ik_Q s}\rangle$,

$$n(k_Q)=\int dk_x \int dk_y n(k_x,k_y,k_z) = \frac{1}{2\pi}\int_{-\infty}^{\infty} ds\, e^{ik_Q s}\langle e^{-ik_Q s}\rangle , \quad (48)$$

where the z axis is defined along Q. To prove these relations, we can introduce a δ function into $n(k_Q)$ and use the exponential representation of it,

$$n(k_Q)=\int dk' n(k')\delta(k_Q-k') = \frac{1}{2\pi}\int ds\, e^{-ik_Q s}\int dk' n(k') e^{ik's} = \frac{1}{2\pi}\int ds\, e^{-ik_Q s}\langle e^{ik's}\rangle , \quad (49)$$

so that $n^{-1}\rho_1(s\hat{\mathbf{Q}},0)=\langle e^{-ik_Q s}\rangle$ as noted.

In (3) and (43), we expanded $\langle e^{-ik_Q s}\rangle$ in powers of $s=qt$ and retained terms up to t^4. This corresponds to selecting a model for $\rho_1(s\hat{\mathbf{Q}},0)$ of

$$n^{-1}\rho_1(s\hat{\mathbf{Q}},0)=e^{-(1/2)k_0^2 s^2}\left[1+\frac{1}{4!}\delta k_0^4 s^4\right] , \quad (50)$$

where $k_0^2=\langle k_Q^2\rangle$ and $\delta=[\langle k_Q^4\rangle-3\langle k_Q^2\rangle^2]/k_0^4$ is the excess of the distribution and, using the Fourier transform (48), we obtain $n(k_Q)$ and the three-dimensional $n(\mathbf{k})$ as

$$n(\mathbf{k})=(2\pi k_0^2)^{-3/2}e^{-k^2/2k_0^2}\times\left[1+\frac{\delta}{8}\left[5-10\frac{k^2}{3k_0^2}+\frac{k^4}{3k_0^4}\right]\right] . \quad (51)$$

For isotropic fluids, once $n(k_Q)$ is determined, $n(\mathbf{k})$ can be found with no loss of information.

Expansions such as (4) are expansions of the OBDM valid for small s. They imply a model for $n(k_Q)$, which can be obtained directly by Fourier transform. We expect this model and method to work well in those materials where the OBDM is short ranged, where $n(\mathbf{k})$ is indeed nearly Gaussian. For example, we do not expect this method to be useful in superfluid ^{4}He where $\rho_1(s\hat{\mathbf{Q}},0)$ has tails extending to large s due to the condensate.

In Fig. 2, we compare the present $n(\mathbf{k})$ in (51) using the parameters listed in Table II with the path-integral Monte Carlo (PIMC) evaluations of Ceperley and Pollock.[40] Since the observed and PIMC kinetic energies in Table II agree well, the Gaussian component of the observed and calculated $n(\mathbf{k})$ agree well. The PIMC $n(\mathbf{k})$ lies above the extracted $n(\mathbf{k})$ at low k but not outside the combined error of k_0 and δ. As dashed lines in Fig. 2, we show (51) for $\delta=0$ (pure Gaussian) and $\delta=1.0$. These comparisons show that $n(\mathbf{k})$ differs significantly from a Gaussian in normal liquid ^{4}He and that $n(\mathbf{k})$, when represented by (51), must have a large positive excess. The $n(\mathbf{k})$ at low k is clearly sensitive to δ.

TABLE I. The parameters μ_2 to μ_6 for normal ^{4}He at $T=2.5$ K obtained by fitting the additive approach (AA) (45) and the convolution approach (CA) (21) to data at $Q=8$ and 14 Å^{-1}. In the AA, terms out to $S_2(Q,\omega)$ only in (45) were retained so that only μ_2, μ_3, and μ_4 are determined.

Q (Å^{-1})	T (K)	Fit	μ_2/Q^2 (meV/Å)^2	μ_3/Q^2 (meV^3 Å^2)	μ_4/Q^4 (meV Å)^4	μ_5/Q^4 (meV^5 Å^4)	μ_6/Q^6 (meV Å)^6
8	2.5	AA	0.85	3.2	0.30		
8	2.5	CA	0.89	2.8	0.28	3.1	0.6
14	2.5	AA	0.97	3.0	0.52		
14	2.5	CA	0.97	3.2	0.54	5.0	0.6

TABLE II. Parameters (from Ref. 41) for the momentum distribution $n(\mathbf{k})$ in (51) for normal ^{4}He obtained from α_2 and α_4; $k_0^2 \equiv \langle k_Q^2 \rangle = (\hbar^2/M)^{-2}(\alpha_2/Q^2)$, $\delta \equiv [\langle k_Q^4 \rangle - 3\langle k_Q^2 \rangle^2]/\langle k_Q^2 \rangle^2 = \alpha_4/\alpha_2^2$, and $(\hbar^2/M) = 1.0442$ meV Å^2 $= 12.12$ K Å^2 for ^{4}He. The corresponding kinetic energy $\langle K \rangle$ and $\langle K \rangle$ calculated by Ceperley and Pollock (PIMC) (Ref. 43) are also listed.

α_2/Q^2 (meV Å)^2	k_0^2 (Å^{-2})	α_4/Q^4 (meV Å)^4	δ	$\langle$K$\rangle$ (K)	$\langle$K$\rangle$ (PIMC) (K)
0.94±0.03	0.86	0.56±0.06	0.63	15.7±0.5	16.0

Our purpose here has been illustrative. When applied systematically, we believe this technique offers a straightforward method to extract $n(\mathbf{k})$. This is the topic of a future paper.

C. Convolution approach

To obtain FS effects in the form of a FS broadening function, $R(Q,\omega)$ we follow the method of Secs. III and IV directly. That is, we fit (41) directly to the observed data, properly convoluted as in (47), with the μ_n treated as free parameters. Since β_4 will be positive, it is necessary to keep terms up to μ_6 in (41) so that the $R(Q,t)$ can be Fourier transformed. We found μ_6 could be determined within a factor of 2. The μ_6 affects $R(Q,\omega)$ chiefly at high ω. We assumed that $\alpha_6 = 0$ and assigned the whole of μ_6 to β_6. The chief role of μ_6 is to ensure convergence of the Fourier transforms, and, since μ_6 could not be well determined, we simply kept μ_6 at a small value ($\mu_6 = 50\,Q^4$ meV^6).

In the lower half of Fig. 1 we show a fit of (41) to the observed scattering intensity at $Q = 8$ Å^{-1} in normal ^{4}He at $T = 2.5$ K. The parameters μ_n up to $n = 5$ were retained, and the fitted values are listed in Table I. Clearly, it is again possible to get a good fit. Also the parameters obtained using the CA (21) and the AA (45) are consistent. In Fig. 3 we show a CA fit at $Q = 14$ Å^{-1}. The values for μ_n at $Q = 14$ Å^{-1} represent the incoherent limit well, although the μ_5 obtained at $Q = 14.0$ Å^{-1} is somewhat higher than the average (compare Tables I and III). Fit statistics are discussed in Appendix C. If we make again the approximation (43) for $S_{\rm IA}$ (to obtain a convergent Fourier transform), it is clear that the CA method above will yield the same $n(\mathbf{k})$ as the AA of Sec. V A.

In Fig. 4, we show $R(Q,\omega)$ obtained from (26) and (27) using the CA parameters listed in Table I and $\alpha_2/Q^2 = 0.965$ meV^2 Å^2 and $\alpha_4/Q^4 = 0.53$ (meV Å)^4 from Table II. That is, Fig. 4 is the Fourier transform of

$$R(Q,t) = \exp\left[-\frac{1}{2}(\mu_2-\alpha_2)t^2 + \frac{i}{3!}\mu_3 t^3 + \frac{1}{4!}(\mu_4-\alpha_4)t^4 - \frac{i}{5!}\mu_5 t^5 - \frac{1}{6!}\mu_6 t^6\right]. \tag{52}$$

Although $Q = 8$ Å^{-1} is not quite in the incoherent limit, $S(Q) = 1.0$ accurately so that (37) reduces to (52). In the incoherent limit, $\mu_2 = \alpha_2$. The peak position of $R(Q,\omega)$ is

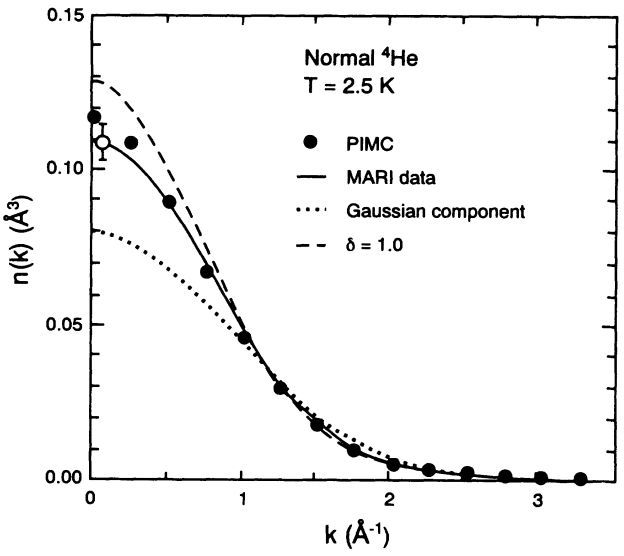

FIG. 2. Momentum distribution $n(\mathbf{k})$ in normal ^{4}He at $T = 2.5$ K. Solid line is $n(\mathbf{k})$ extracted by fitting the additive approach (45) and the full function (41) to the data of Andersen *et al.* (Ref. 40) at several Q values. The extracted $n(\mathbf{k})$ is given by (53) with $k_0^2 = 0.86$ Å^{-1}, $\delta = 0.63$ (see Table II). (The corresponding kinetic energy is $\langle K \rangle = 15.7 \pm 0.4$ K). The dotted and dashed line is (53) with the same $\langle K \rangle$ but $\delta = 0$ and $\delta = 1.0$, respectively. The solid dots are the PIMC $n(\mathbf{k})$ calculated at $T = 2.5$ K by Ceperley and Pollock (Ref. 43) ($\langle K \rangle = 16.0$ K).

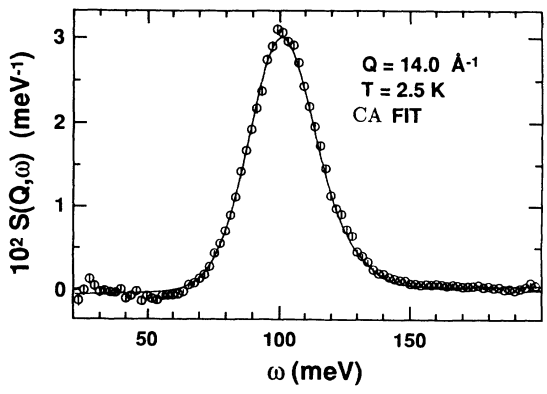

FIG. 3. Convolution approach (CA) (41) fitted to data of Andersen (Ref. 39) at $Q = 14$ Å^{-1} and $T = 2.5$ K. The instrument resolution was represented by a Gaussian with $\sigma_I = 3.4$ meV, and the resulting fitting parameters, μ_{2i}–μ_{6i}, are listed in Table I.

TABLE III. Average values of the parameters μ_{2i} to μ_{6i} in the incoherent limit found by fitting the AA and the CA methods to data at several Q values in normal ^{4}He at $T=2.5$ K. The parameters α_2 and α_4 appearing in the IA (43) are obtained as $\alpha_2=\mu_{2i}$ and $\alpha_4=\mu_{4i}$ using these averages. As noted in the text, μ_6 was fixed at $\mu_6/Q^4=50$ (meV Å^{-2})^6.

μ_{2i}/Q^2 (meV Å)^2	μ_{3i}/Q^2 (meV^3 Å^2)	μ_{4i}/Q^4 (meV Å)^4	μ_{5i}/Q^4 (meV^5 Å^4)	μ_{6i}/Q^4 (meV Å^{-2})^6
0.94±0.03	2.5±0.5	0.56±0.06	2.7±0.5	50

determined chiefly by μ_3. There is some cancellation between the effects of μ_3 and μ_5. A large μ_3 unbalanced by μ_5 leads to large amplitude oscillations in $R(Q,\omega)$ at low ω. A large μ_5 unbalanced by μ_3 leads to oscillations in $R(Q,\omega)$ at high ω. Some oscillation is clearly needed to fulfill the moment relations (39). The properties of $R(Q,\omega)$ will be discussed further in a future paper.[35]

As noted above, we had difficulty determining the FS contribution β_4 to μ_4. The μ_4 was dominated by the IA term α_4. To estimate $\beta_4=\mu_4-\alpha_4$, we expanded the IA in the form (43) and used this expanded form of $S(Q,t)$ in (21) to give

$$S(Q,t)=S(Q)e^{-i\omega_R' t}\left[1+\frac{1}{4!}\alpha_4 t^4\right] \times\exp\left[-\frac{1}{2}\mu_2 t^2+\frac{i}{3!}\mu_3 t^3+\frac{1}{4!}(\mu_4-\alpha_4)t^4 -\frac{i}{5!}\mu_5 t^5-\frac{1}{6!}\mu_6 t^6\right] . \quad (53)$$

The α_4 was not further adjusted, which corresponds to keeping $n(\mathbf{k})$ fixed. The μ_2 to μ_6 were then found by fitting to experiment and $R(Q,t)$ is obtained directly from (15) or (26). At $Q=8$ Å^{-1} we found similar parameters to those in Table I with $(\mu_4-\alpha_4)/Q^4=0.28$ (meV Å)^4. Thus, $\beta_4=\mu_{4i}-\alpha$, the final-state contribution to μ_{4i}, is small, and this procedure provides some estimate of it.

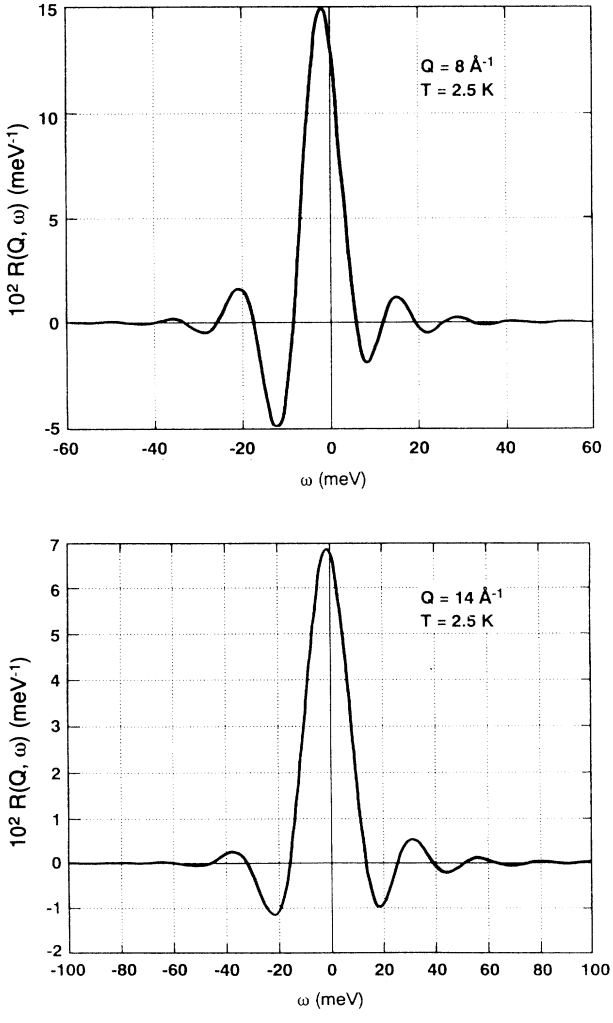

FIG. 4. Final-state broadening function, $R(Q,\omega)$, obtained at $Q=8$ and 14 Å^{-1} in normal ^{4}He at $T=2.5$ K. $R(Q,\omega)$ is obtained from (52) and (9) using the parameters listed in Tables I and II.

VI. APPLICATIONS FOR NON-GAUSSIAN $n(\mathbf{k})$: SUPERFLUID ^{4}He

In Sec. V, we fitted the functions (41) and (45) to data in normal ^{4}He and found the momentum distribution $n(\mathbf{k})$ given by (51). As discussed in Sec. V B, this corresponds to selecting a particular model for $n(\mathbf{k})$ in which $n(\mathbf{k})$ is represented by its second and fourth moments: k_0^2 and $\delta=\alpha_4/\alpha_2^2$.

To apply the method to a system in which $n(\mathbf{k})$ is not well represented by its second and fourth moments alone, we may introduce an appropriate model $n(\mathbf{k})$. The IA for this model $n(\mathbf{k})$ is then calculated from the standard expression (1),

$$S_{\rm IA}(Q,\omega)=\int d\mathbf{k}\, n(\mathbf{k})\delta(\omega-\omega_R-qk_Q) . \quad (54)$$

The model $n(\mathbf{k})$ may contain parameters to be determined by fitting to experiment, or it could be a calculated $n(\mathbf{k})$ that we wish to test. The $S_{\rm IA}(Q,\omega)$ is then convoluted with the $R(Q,\omega)$ given by (9) and (26) to give $S(Q,\omega)$ as

$$S(Q,\omega)=\int d\omega' S_{\rm IA}(Q,\omega')R(Q,\omega-\omega') \quad (55)$$

in the usual way. The $S(Q,\omega)$ now depends on the parameters in $n(\mathbf{k})$ and the β_2 to β_6 in $R(Q,\omega)$. There will be consistency relations that will reduce the number of free parameters to be determined by fitting to experiment as discussed below. The resulting $S(Q,\omega)$ can be fitted with the aim of testing $n(\mathbf{k})$ or determining adjustable

parameters in $n(\mathbf{k})$, or $n(\mathbf{k})$ can be taken as given and the parameters β_n in $R(Q,t)$ can be refined. This method can be applied to any fluid or solid. To illustrate, we apply the procedure to superfluid ^{4}He.

A. Convolution approach in superfluid ^{4}He

Andersen *et al.*[41] have measured the coherent $S(Q,\omega)$ in superfluid ^{4}He at $T=1.42$ K in the range $5 \lesssim Q \lesssim 10$ Å^{-1}. They also fitted the additive approach (AA) to this data as noted above for normal ^{4}He. We have fitted the convolution approach (CA) to Andersen's data at several Q values. The fits were good and the CA (41) therefore represents the data well. The resulting parameters $\mu_n(Q)$ oscillate with Q in the range $5 \lesssim Q \lesssim 10$ Å^{-1} as in normal ^{4}He and the CA can be used to display the oscillations in the peak position and width of $S(Q,\omega)$ with Q accurately. The oscillations continue up to $Q \simeq 10$ Å^{-1}. Since the data did not continue past $Q=10$ Å^{-1}, we were not able to get precise values of the incoherent limit of the μ_{ni} [by averaging the $\mu_{ni}(Q)$ at several Q values at high Q as done above for normal ^{4}He].

The chief change in the CA fit in going from $T=2.5$ to 1.42 K is a drop in μ_2/Q^2 by approximately 0.1 (meV Å)^2. This is shown in Fig. 5, where μ_2/Q^2 at the two temperatures is compared. In the incoherent limit, where $\mu_2=\alpha_2$, this corresponds to a drop in kinetic energy of approximately 1.8 K. At $T=2.5$ and 1.42 K, Ceperley and Pollock[43] find $\langle K \rangle = 16.0$ and $\langle K \rangle = 14.3$ K, respectively, a drop of 1.7 K. Otherwise, the higher-order parameters μ_3, μ_4, and μ_5 are unchanged within the precision they are determined. This suggests that the FS resolution function $R(Q,\omega)$ (which depends chiefly on μ_3 and μ_5) changes little or not at all in going from normal to superfluid ^{4}He. $R(Q,\omega)$ depends on the interaction between the atoms, especially at short range, and we expect these to change little with temperature.

Thus, in going from normal to superfluid ^{4}He we find a clear drop in μ_2 (the kinetic energy) but the higher μ_n unchanged within precision. We now introduce a model $n(\mathbf{k})$ having a condensate. We essentially attribute the change in μ_2 to the appearance of a condensate and thereby determine the condensate in the model.

B. Model momentum distribution

To describe the momentum distribution $n(\mathbf{k})$ in superfluid ^{4}He, we introduce the following model:

$$n(\mathbf{k})=n_0\delta(\mathbf{k})+n_0[f(\mathbf{k})+A_i f_i(\mathbf{k})]+A_1 n^*(\mathbf{k}) , \quad (56)$$

where

$$n_0 f(\mathbf{k})=\left[\frac{n_0 Mc}{(2\pi)^3 2\hbar n}\right]\frac{1}{k}[2n_B(ck)+1]e^{-k^2/2k_c^2} , \quad (57)$$

$$f_i(\mathbf{k})=(2\pi k_i^2)^{-3/2}e^{-k^2/2k_i^2} , \quad (58)$$

$$n^*(\mathbf{k})=(2\pi k_0^2)^{-3/2}e^{-k^2/2k_0^2}\left\{1+\frac{\delta}{8}\left[5-\frac{10}{3}\left[\frac{k}{k_0}\right]^2+\frac{1}{3}\left[\frac{k}{k_0}\right]^4\right]\right\} . \quad (59)$$

Here $n_0\delta(\mathbf{k})$ is the condensate contribution and $n_0[f(\mathbf{k})+A_i f_i(\mathbf{k})]$ is the increase in $n(\mathbf{k})$, chiefly at small k, due to the coupling between the single particle and density response of the superfluid via the condensate. The term, $n_0 f(\mathbf{k})$, arises from the sharp peak (phonon peak) in $S(Q,\omega)$ at low $Q=k$, the joint-density–single-particle response (see Appendix C). How this terminates at higher k is not well known, and we have introduced the cutoff, $e^{-k^2/2k_c^2}$. The term $n_0 f_i(\mathbf{k})$, due to the incoherent, broad response at higher k, is not well known,

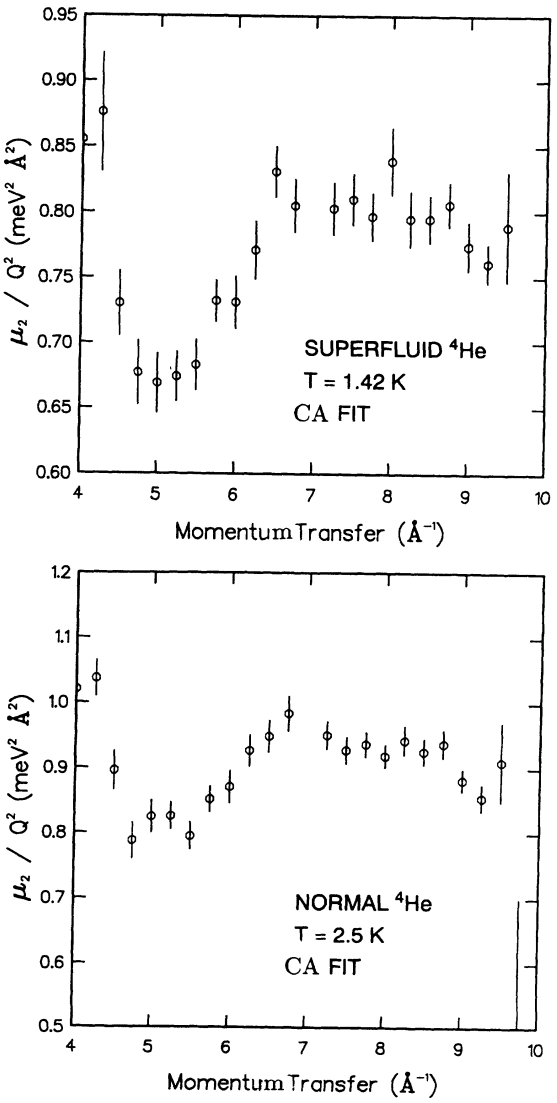

FIG. 5. Values of μ_2/Q^2 at $T=2.5$ and 1.42 K obtained by fitting the CA (41) to data of Andersen (Ref. 39) using a Gaussian instrument resolution function with $\sigma_I=1.27$ meV.

and we represent this by a Gaussian. The $n^*(\mathbf{k})$ is the momentum distribution over states above the condensate, which we assume is the same as in normal ^{4}He. Thus we use (51) for $n^*(\mathbf{k})$ and take k_0^2 and δ as determined above in Sec. V for normal ^{4}He at 2.5 K (see Table II). The k_i we take arbitrarily as $k_i=2$ Å^{-1}, a long wave vector. The determination of k_c is discussed below. With these choices, there remain three free parameters in $n(\mathbf{k})$: n_0, A_1, and A_i. This choice of $n(\mathbf{k})$ is very much in the spirit proposed by Woods, Svensson, and coworkers,[10–13] the sum of a condensate-induced contribution plus $n^*(\mathbf{k})$ describing the states above the condensate.

The $n(\mathbf{k})$ should be normalized and the moments $\langle k_Q^n \rangle$ calculated with $n(\mathbf{k})$ should be consistent with the observed moments, α_n, in the superfluid. Up to $\langle k_Q^2 \rangle$ this gives,

$$\int d\mathbf{k}\, n(\mathbf{k})=1 \ , \tag{60a}$$

$$\frac{1}{\langle k_Q^2 \rangle_N}\int d\mathbf{k}\, n(\mathbf{k})k_Q^2=\frac{\langle k_Q^2 \rangle_S}{\langle k_Q^2 \rangle_N}=\frac{\langle K \rangle_S}{\langle K \rangle_N}\equiv R \ . \tag{60b}$$

We have divided (60b) by the second moment in the normal fluid $\langle k_Q^2 \rangle_N$ so that the result can be written as the ratio of the kinetic energies in the superfluid and normal phases (see Table IV). Higher-moment relations can also be used.

Substituting $n(\mathbf{k})$ from (56) into (60) and carrying out the integrations indicated, we obtain ($k_0^2 \equiv \langle k_Q^2 \rangle_N$),

$$n_0[1+I_0+A_i]+A_1=1 \ , \tag{61a}$$

$$n_0 I_2' + A_i(k_i/k_0)^2 + A_1 = R \ , \tag{61b}$$

where $I_n=\int d\mathbf{k}\, f(\mathbf{k})k_Q^n$ and $I_2'=I_2/k_0^2$. The I_n depend on k_c, but otherwise can be calculated directly. Equations (61) provides two relations to eliminate two of the three free parameters, n_0, A_1, and A_i, in $n(\mathbf{k})$. In the present application, we have determined R from the ratio of the kinetic energies; $\langle K \rangle_N=16.0$ and $\langle K \rangle_S=14.3$ K as calculated by Ceperley and Pollock,[43] which gives $R=0.893$. Preferably we should use a $\langle K \rangle$ determined from $S(Q,\omega)$ itself in the incoherent limit. This will be done in subsequent work using new data at higher Q values in the superfluid, which have become available. We now implement this method to determine n_0 beginning with a simplified version obtained by taking $A_i=0$.

C. The condensate

1. Simple model

We now determine the condensate $n_0(T)$ by substituting the model $n(\mathbf{k})$ of (56) into the IA (54) and fitting the resulting $S(Q,\omega)$ to observed data at $Q=8$ Å^{-1}. We begin with a simple model obtained by setting $A_i=0$. Equations (61) then reduce to

$$n_0[1+I_0]+A_1=1 \ , \tag{62a}$$

$$n_0 I_2' + A_1 = R \ . \tag{62b}$$

These can be solved immediately to get n_0 in terms of R, $n_0=(1-R)/[1+I_0-I_2']$, independently of any fit (see Table V). This solution is essentially the method proposed by Sears[44] to obtain $n_0(T)$ from the kinetic energy, here from the ratio $R=\langle K \rangle_S/\langle K \rangle_N=0.893$. In this application we have included $f(\mathbf{k})$ so that n_0 depends upon $f(\mathbf{k})$, and particularly on the cut off k_c as well as on R. In Table VI we list I_0 and I_2' for $0.3\le k_c\le 0.7$ Å^{-1} and the corresponding n_0 obtained by solving (62). We see that n_0 varies between $n_0=0.09$ and 0.07 between these limits. A very reasonable choice of k_c is $k_c=0.5$ Å^{-1}. The $f(\mathbf{k})$ in (57) is obtained assuming linear phonon dispersion $\omega=ck$ in the phonon-roton excitations. Linearity is observed to cease at $k\approx 0.7$ Å^{-1}. Thus, we should cut off $f(k)$ at least by $k\approx 0.7$ Å^{-1}, which will be the case if $k_c=0.5$ Å^{-1}. Certainly k_c must lie within the range shown in Table I. We take $k_c=0.5$ giving the result $n_0=0.08\pm 0.01$. For $k_c=0$, $n_0=0.11$ which is the result that would be obtained by the Sears[45] method with the present $\langle K \rangle$ values.

With $n_0=0.08$ and A_1 determined by normalization, $n(\mathbf{k})$ in (56) is completely fixed. We fit the resulting $S(Q,\omega)$ to experiment by adjusting the parameters μ_2 to μ_5, with μ_6 set at $\mu_6=50/Q^4$. The fit at $Q=8$ Å^{-1} and $T=1.42$ K is shown in the upper part of Fig. 6. We see that the fit is good. The chief discrepancy is that the fitted $S(Q,\omega)$ is too low in the peak region, which suggests that n_0 is somewhat too small. That is, if we substitute $n_0\delta(\mathbf{k})$ into (54) and (55), we obtain a contribution to $S(Q,\omega)$ of

$$n_0 R(Q,\omega-\omega_R) \ , \tag{63}$$

which clearly contributes chiefly at $\omega=\omega_R$, where $R(Q,\omega-\omega_R)$ peaks. The $n_0 f(\mathbf{k})$ is also sufficiently narrow compared to $R(Q,\omega)$ that it contributes in the same way as $n_0\delta(\mathbf{k})$. Thus, the discrepancy in the peak region in the upper part of Fig. 6 suggests $n_0=0.08$ is too small. To test this we have arbitrarily increased n_0 to $n_0=0.10$ and 0.12, while keeping all other parameters constant except A_1, which was adjusted to maintain normalization of $n(\mathbf{k})$ in (56). The corresponding fits are shown in the middle and lower frames of Fig. 6. The value $n_0=0.10$ fits best in the peak region.

There is a second interesting effect when n_0 is increased. In the wings of $S(Q,\omega)$, e.g., $40\lesssim\omega\lesssim 60$ meV,

TABLE IV. Parameters used in $n(\mathbf{k})$ in (56) and (60) for superfluid ^{4}He at $T=1.42$ K.

α_2^N/Q^2 (meV Å)^2	k_0^2 (Å^{-2})	α_4^N/Q^4 (meV Å)^4	δ	α_2^S/Q^2 (meV Å)^2	α_4^S/Q^4 (meV Å)^4	k_c (Å^{-1})	k_i (Å^{-1})
0.94	0.86	0.56	0.63	0.85	0.56	0.5	2.0

TABLE V. Parameters μ_2 to μ_6 obtained by fitting $S(Q,\omega)$ in (55) with the model $n(\mathbf{k})$ of (56) in the impulse approximation to data at $Q=8$ Å^{-1} in superfluid ^{4}He at $T=1.42$ K. In the model $n(\mathbf{k})$, $A_i=0$ [simple model (SM)]. In SM(1), all μ_n are adjusted to fit the data (upper part of Fig. 6). In SM(2) μ_2 is fixed at $\mu_2=\alpha_2^S$ to simulate the incoherent limit (see Fig. 8). A Gaussian instrument resolution function was used with $\sigma_I=1.27$ meV.

Model		T	μ_2/Q^2	μ_3/Q^2	μ_4/Q^4	μ_5/Q^4	μ_6/Q^4
$n(\mathbf{k})$	SM(1)	1.42 K	0.897	2.95	0.697	3.23	50
$n(\mathbf{k})$	SM(2)	1.42 K	0.858	2.94	0.642	3.31	50

the fitted line oscillates around the observed data. These oscillations originate from the oscillations in $R(Q,\omega)$ at larger ω (see Fig. 7) and enter $S(Q,\omega)$ through the term (63). Thus the contribution from n_0 can be seen away from the peak region due to $R(Q,\omega)$. A refitting would result in a reduction in μ_3 and μ_5, which would reduce the oscillations. They are clearly not in the data. However, a refitting does not remove the oscillations entirely, as can be seen in the fitted $n_0=0.08$ case; there is a competition between fitting in the peak and in the wings, which leaves some oscillation in the fitted function. Finally, a value $n_0=0.10$ and the simple model is just compatible with the ratio of the kinetic energies $R=14.3/16.0$ if we assign an error of ± 0.03 to each kinetic energy.

When a condensate is inserted, there is some trade off between the value of μ_2, μ_4, and n_0. To illustrate this, we show a fit of $S(Q,\omega)$ at $Q=8$ Å^{-1} with μ_2 arbitrarily fixed at $\mu_2=\alpha_2$, the incoherent limit, in Fig. 8. We expect this to be a lower limit of the μ_2, given there is a condensate added. In this case $n_0=0.08$ gives a good fit in the peak region. Similarly, if we arbitrarily adjust μ_4 downward to the incoherent limit, $n_0=0.12$ gives the best fit.

2. *Full model*

We have implemented the full model by treating A_i as a parameter and calculating n_0 and A_1 using (61). The best fit gives $A_i=0.03$ and $n_0=0.11$. The fit is comparable to the upper part of Fig. 6 but fits better in the peak region. The broad component is clearly small and n_0 is consistent with the determinations above.

We conclude with the following comments. Firstly, it is possible to obtain a reasonable fit to the data without introducing a condensate. In this sense there is no unique signature of the condensate at these Q values. However, the fit requires a large drop in the kinetic energy, which is inconsistent with the temperature-independent $\langle K \rangle$ in normal ^{4}He unless there is some unusual feature, i.e., a condensate. This point is discussed more fully by Sears.[44] If we ascribe the drop in kinetic energy to movement of atoms into the condensate, which is the basis of the model $n(\mathbf{k})$ in (56), then an $n_0=0.10\pm0.03$ is obtained for

TABLE VI. Moments $I_n=\int d\mathbf{k}\, f(\mathbf{k})k_Q^n$ of the function $f(\mathbf{k})$ defined in (57) as a function of the cutoff parameter k_c; $I_2'=I_2/k_0^2$. The n_0 is the corresponding condensate fraction obtained by solving (62) ($A_i=0$) showing the sensitivity of n_0 to k_c.

k_c (Å^{-1})	I_0	I_2 (Å^{-2})	I_2'	n_0
0.3	0.19	0.01	0.01	0.093
0.4	0.31	0.03	0.03	0.086
0.5	0.47	0.07	0.08	0.078
0.6	0.66	0.15	0.16	0.073
0.7	0.89	0.28	0.30	0.069

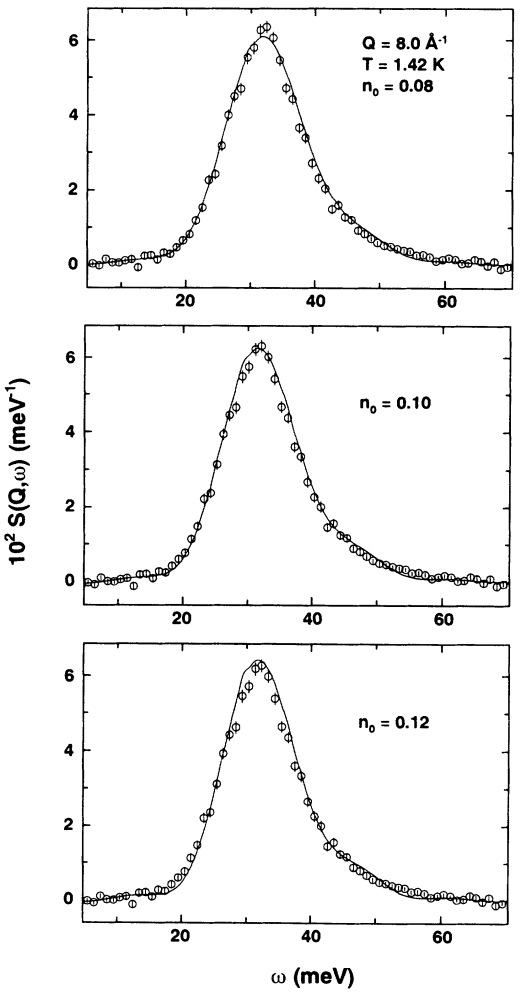

FIG. 6. Fit of $S(Q,\omega)$ using the model $n(\mathbf{k})$ given by (56) with $A_i=0$ (simple model) to data of Andersen (Ref. 39) at $Q=8$ Å^{-1} and $T=1.42$ K. The upper graph shows results obtained by solving (62) directly, which gives $n_0=0.08$ in the simple model. The middle and lower graphs show results for n_0 increased to $n_0=0.10$ and $n_0=0.12$, respectively, with all other parameters held constant.

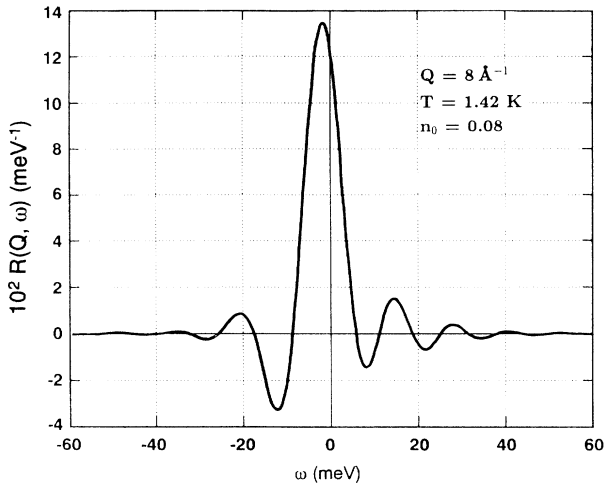

FIG. 7. Final-state (FS) resolution function $R(Q,\omega)$ obtained at $Q=8$ Å^{-1} using the model $n(\mathbf{k})$ in (58) with $A_i=0$ and $n_0=0.08$ in superfluid ^{4}He at $T=1.42$ K. The fitted parameters μ_2 to μ_6 are listed in Table V as SM(1).

reasonable values of the kinetic energy and fits to the data. The uncertainty in the fit and in $R(Q,\omega-\omega_R)$ leads to an uncertainty in n_0 of approximately ± 0.02. The uncertainty in the cut off of $f(\mathbf{k})$ adds an error of ± 0.01 to n_0.

We believe this method can be improved when applied to several Q values in the incoherent regime. For example, using the scaling method discussed in the next section an accurate $R(Q,\omega)$ obtained from data at several Q values can be determined. This, combined with accurate kinetic energies and the model $n(\mathbf{k})$ in (56), can be used to determine $n_0(T)$ more reliably and perhaps other character of $n(\mathbf{k})$. This work is in progress.[35] The unique signature of the condensate is seen in other measurements, such as the existence of the roton.

VII. SCALING OF $R(Q,\omega)$

In the incoherent limit, the present FS broadening function $R(Q,\omega)$ obtained in the form (15) at one Q, may be scaled to any Q. This is readily done using the usual y scaling variable, $y=q^{-1}(\omega-\omega_R)$, and $s=qt$ discussed above, where $q=\hbar Q/M$. Indeed, the present method for obtaining $n(\mathbf{k})$ and $R(Q,\omega)$ may be transparently presented in these variables if the coherent regime is not needed.

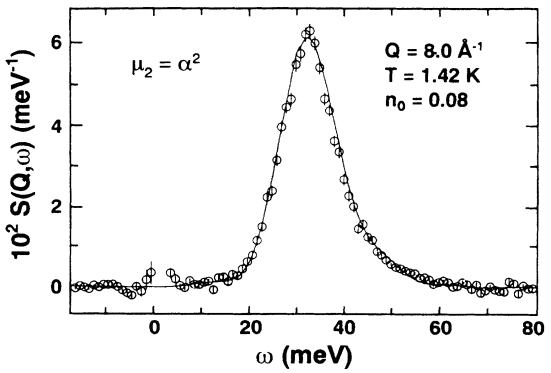

FIG. 8. Fit of $S(Q,\omega)$ as in Fig. 6 with μ_2 fixed at $\mu_2=\alpha_2^S$. The parameters are listed in Table V as SM(2).

The IA depends only on y rather than Q and ω separately. Introducing y into (1), we have

$$J_{\rm IA}(y)\equiv qS_{\rm IA}(Q,\omega)=\int d\mathbf{k}\, n(\mathbf{k})\delta(y-k_Q) =n(y)\ , \tag{64}$$

where $J_{\rm IA}(y)=n(k_Q)$ follows by carrying out the integration $d\mathbf{k}=dk_x dk_y dk_z$ in (64) as in (48). $J_{\rm IA}(y)$ is the longitudinal-momentum-distribution function, and y is the momentum variable along Q. The corresponding intermediate-scattering function, from (3) or (48), is $J_{\rm IA}(s)=\langle e^{-ik_Q s}\rangle$. Although $S(Q,\omega)$ and $R(Q,\omega)$ do not scale as y, it is usual to define similarly scaled functions,

$$J(Q,y)\equiv qS(Q,\omega),\quad R(Q,y)\equiv qR(Q,\omega-\omega_R)\ , \tag{65}$$

so that (8) becomes

$$J(Q,y)=\int dy' J_{\rm IA}(y-y')R(Q,y')\ . \tag{66}$$

The corresponding intermediate scattering function, from (11), is

$$J(Q,s)=\left\langle T_s \exp\left[-i\int_0^s ds' k_Q(s')\right]\right\rangle\ .$$

Using the Fourier transform (9), we have

$$R(Q,y)=\frac{q}{2\pi}\int dt\, e^{-i(\omega-\omega_R)t}R(Q,t) =\frac{1}{2\pi}\int ds\, e^{iys}R(Q,s)\ . \tag{67}$$

The expansions (4) and (12) are

$$J_{\rm IA}(s)=\langle e^{-ik_Q s}\rangle=\exp\left[\sum_{n=2}^{\infty}\frac{(-is)^n}{n!}\bar{\alpha}_n\right]\quad (n\ \text{even}) \tag{68}$$

with $\bar{\alpha}_n$ defined in (5) and

$$J(Q,s)=\left\langle T_s \exp\left[-i\int_0^s ds' k_Q(s')\right]\right\rangle =\exp\left[\sum_{n=2}^{\infty}\frac{(-is)^n}{n!}\bar{\mu}_n\right] \tag{69}$$

with $\bar{\mu}_n=\mu_{ni}/q^n$ given by (34), respectively. Using the definition $J(Q,s)=J_{\rm IA}(s)R(Q,s)$, we obtain from (68) and (69),

$$R(Q,s)=\exp\left[\sum_{n=3}^{\infty}\frac{(-is)^n}{n!}\bar{\beta}_n\right]\ , \tag{70}$$

where $\bar{\beta}_n\equiv\bar{\mu}_n-\bar{\alpha}_n$ and, as noted, $\beta_2=0$ and $\bar{\alpha}_n=0$ for odd n.

Using (15) or (70) for $R(Q,s)$, $\bar{\beta}_n=\mu_{ni}/q^n-\bar{\alpha}_n$, and substituting (34) for μ_{ni}, we obtain

$$R(Q,s)=\exp\left[\frac{i}{3!}\frac{\bar{a}_3}{q}s^3+\frac{1}{4!}\frac{\bar{a}_4}{q^2}s^4-\frac{i}{5!}\left[\frac{\bar{a}_{52}}{q^3}+\frac{\bar{a}_{54}}{q}\right]s^5-\frac{1}{6!}\left[\frac{\bar{a}_{62}}{q^4}+\frac{\bar{a}_{64}}{q^2}\right]s^6\right] \tag{71}$$

up to $0(s^6)$, where the $\bar{a}$ are independent of Q. In liquid ^{4}He, we found $\bar{a}_{52}/q^3$ was negligible and $\bar{a}_4/q^2$ was small. Thus, $R(Q,s)$ depends chiefly on $\bar{a}_3$ and $\bar{a}_{54}$. We were not able to determine $\beta_6=\bar{a}_{62}q^2+\bar{a}_{64}q^4$, and we chose β_6 so that the Fourier transform converged, i.e., $R(Q,s)\to 0$ beyond physically realizable values of s. We now show that $s\lesssim 5$ Å^{-1} in helium, independent of Q.

The Fourier transform of $J(Q,y)$ is

$$J(Q,y)=\frac{1}{2\pi}\int ds\, e^{iys}\langle e^{-ik_Q s}\rangle R(Q,s)\ . \tag{72}$$

The $J_{\rm IA}(s)=\langle e^{-ik_Q s}\rangle\approx e^{-s^2/s_\tau^2}$ goes to zero when the struck particle travels a distance $s\sim s_\tau=\sqrt{2}/\langle k_Q^2\rangle\simeq 1.5$ Å^{-1}. Thus s values $s\gg s_\tau$ contribute little to (72) and $R(Q,s)$ for $s\gg s_\tau$ is not important. We found that $\beta_6=\bar{a}_{64}q^4+\bar{a}_{62}q^2\simeq 50Q^4$ meV^6 in (71) reduces $R(Q,s)$ to zero for $s\simeq 4$ Å and provides a reasonable cut off. Any reasonable cut off will do in the Q range considered here. Equations (67) and (71) provide a universal value of $R(Q,y)$ in any system once the coefficients $\bar{a}_n$ are determined.

VIII. SUMMARY

We have proposed functions, (41) and (45), which represent $S(Q,\omega)$ well at intermediate and high Q and which have a physical basis. The coefficients μ_n reach the incoherent limit at $Q\simeq 10$ Å^{-1}, a value higher than anticipated from $S(Q)$.

Using (41) and (45), we have extracted the momentum distribution and final-state broadening function in normal ^{4}He at $T=2.5$ K. The $n(\mathbf{k})$ deviates significantly from a Gaussian and agrees quite well with PIMC values. The Gaussian component of $n(\mathbf{k})$ provides a kinetic energy, which also agrees with the PIMC value. The $R(Q,\omega)$ shows the expected form with oscillations at high ω. It is independent of T between $T=2.5$ and 1.42 K within the error of determination, i.e., the $R(Q,\omega)$ obtained at $T=1.42$ K using the CA is the same as that in Fig. 4 except for small deviations in the wings. The method can be generalized to use a model $n(\mathbf{k})$. Introducing a model $n(\mathbf{k})$ having a condensate we find a value of $n_0=0.10\pm 0.03$ in superfluid ^{4}He at $T=1.42$ K. This value can be made more reliable when data in the incoherent regime is used, which is the topic of a future paper.[35] A preliminary report of those results has been made.[46] The method is quite general and can be applied to any solid, fluid, or mixture.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge close collaboration with W. G. Stirling, K. H. Andersen, and R. T. Azuah in developing this fitting method.

APPENDIX A

In this appendix we derive relation (18) for the coherent dynamic structure factor from the standard expression (17). The derivation follows closely similar derivations by Rahman, Singwi, and Sjölander[34] and Gersch, Rodriguez, and Smith.[23] We also show how (18) can be expanded in cumulants to obtain the exponential function relation (21) and (41).

The standard expression (17) is

$$S(Q,\omega)=\frac{1}{N}\sum_i\left\langle e^{-i\mathbf{Q}\cdot\mathbf{r}_i(t)}\sum_j e^{i\mathbf{Q}\cdot\mathbf{r}_j(0)}\right\rangle\ , \tag{A1}$$

where $e^{-i\mathbf{Q}\cdot\mathbf{r}(t)}$ is in the Heisenberg representation,

$$e^{-i\mathbf{Q}\cdot\mathbf{r}_i(t)}=e^{iHt/\hbar}e^{-i\mathbf{Q}\cdot\mathbf{r}_i}e^{-iHt/\hbar}=e^{iHt/\hbar}e^{-i\mathbf{Q}\cdot\mathbf{r}_i}e^{-iHt/\hbar}e^{i\mathbf{Q}\cdot\mathbf{r}_i}e^{-i\mathbf{Q}\cdot\mathbf{r}_i}\ . \tag{A2}$$

We note that $e^{i\mathbf{Q}\cdot\mathbf{r}}$ is a translation operator in momentum, i.e.,

$$e^{-i\mathbf{Q}\cdot\mathbf{r}}f(\mathbf{p},\mathbf{r})e^{i\mathbf{Q}\cdot\mathbf{r}}=f(\mathbf{p}+\hbar\mathbf{Q},\mathbf{r})\ , \tag{A3}$$

where $f(\mathbf{p},\mathbf{r})$ is an arbitrary function of $\mathbf{p}$ and $\mathbf{r}$. Thus, $e^{i\mathbf{Q}\cdot\mathbf{r}_i}$ translates $\mathbf{p}_i$ in H from $\mathbf{p}_i$ to $\mathbf{p}_i+\hbar\mathbf{Q}$, i.e.,

$$e^{-i\mathbf{Q}\cdot\mathbf{r}_i}He^{i\mathbf{Q}\cdot\mathbf{r}_i}=H+\frac{1}{2M}[(\mathbf{p}_i+\hbar\mathbf{Q})^2-p_i^2]=H+\frac{\hbar\mathbf{p}_i\cdot\mathbf{Q}}{M}+\hbar\omega_R \tag{A4}$$

and (A2) becomes

$$e^{-i\omega_R t}e^{iHt/\hbar}e^{-i(H/\hbar+Qv_{iQ})t}e^{-i\mathbf{Q}\cdot\mathbf{r}_i}\ , \tag{A5}$$

where $(\mathbf{p}_i\cdot\mathbf{Q})/M=Qv_{iQ}$. Using the general result,

$$\exp(iAt)\exp[-i(A+B)t]=T_t\exp\left[-i\int_0^t dt'B(t')\right]\ , \tag{A6}$$

where $B(t)=e^{iAt}B(0)e^{-iAt}$ and T_t is the time-ordering operator, we may rewrite (A5) as

$$\exp[-i\mathbf{Q}\cdot\mathbf{r}(t)]=\exp(-i\omega_R t)T_t\exp\left[-iQ\int_0^t dt'v_{iQ}(t')\right]\exp(-i\mathbf{Q}\cdot\mathbf{r}_i)\ . \tag{A7}$$

Substituting (A7) into (A1) then gives

$$S(Q,\omega)=\frac{1}{N}\sum_i\left\langle T_t\exp\left[-iQ\int_0^t dt'v_{iQ}(t')\right]\sum_j\exp[-i\mathbf{Q}\cdot(\mathbf{r}_i-\mathbf{r}_j)]\right\rangle\exp(-i\omega_R t)\ . \tag{A8}$$

Defining $\hat{S}(Q)=\sum_j e^{iQ\cdot(\mathbf{r}_i-\mathbf{r}_j)}$ as the static structure factor operator so that $S(Q)=\langle\hat{S}(Q)\rangle$, we may multiply and divide each term in (A8) by $S(Q)$ to obtain

$$S(Q,t)=S(Q)\exp(-i\omega_R t)\frac{1}{N}\sum_i\left\langle T_t\exp\left[-iQ\int_0^t dt' v_{iQ}(t')\right]\hat{S}(Q)\right\rangle\Big/\langle\hat{S}(Q)\rangle\ . \tag{A9}$$

Each term in (A9) is a normalized expectation value suitable for a cumulant expansion. Also, for a fluid of identical atoms and ignoring surface effects each term is identical and

$$S(Q,t)=S(Q)\exp(-i\omega_R t)\left\langle T_t\exp\left[-iQ\int_0^t dt' v_Q(t')\right]\right\rangle_S\ , \tag{A10}$$

where the expectation value in (A10) is

$$\langle A\rangle_S\equiv\langle A\hat{S}(Q)\rangle/\langle\hat{S}(Q)\rangle\ . \tag{A11}$$

Equation (A10) is the result for $S(Q,t)$ in (18) we wished to derive. We now show how this can be expanded in cumulants.

The standard expression for the cumulant expansion is

$$\ln\langle e^x\rangle=\sum_{n=1}^{\infty}\frac{1}{n!}\mu_n(x)\ , \tag{A12}$$

where $\mu_n(x)$, the cumulants of x, can be derived by expanding the exponential as

$$e^x=1+\sum_{n=1}^{\infty}x^n/n!\equiv 1+y$$

and expanding the logarithm as

$$\ln(1+y)=\sum_{k=1}^{\infty}(-1)^{k-1}y^k/k\ .$$

A similar cumulant expansion of the expectation value in (A10) can be developed by first expanding the exponential in (A10) as

$$\left\langle T_t\exp\left[-iQ\int_0^t dt' v_Q(t')\right]\right\rangle_S =1+\alpha_1(t)+\alpha_2(t)+\alpha_3(t)+\alpha_4(t)+\cdots\ , \tag{A13}$$

where

$$\begin{aligned}\alpha_1&=-iQ\int_0^t dt'\langle v_Q(t')\rangle_S\ ,\\ \alpha_2&=-Q^2\int_0^t dt_1\int_0^{t_1}dt_2\langle v_Q(t_1)v_Q(t_2)\rangle_S\ ,\\ \alpha_n&=(-iQ)^n\int_0^t dt_1\cdots\int_0^{t_{n-1}}dt_n\langle v_Q(t_1)\cdots v_Q(t_n)\rangle_S\ ,\end{aligned} \tag{A14}$$

and secondly expanding the logarithm as indicated above with $y=\alpha_1+\alpha_2+\cdots$. This gives

$$\ln\left\langle T_t\exp\left[-iQ\int_0^t dt' v_Q(t')\right]\right\rangle_S=\sum_{n=1}^{\infty}\mu_n\ , \tag{A15}$$

where

$$\begin{aligned}\mu_1&=\alpha_1\ ,\\ \mu_2&=\alpha_2-\alpha_1^2\ ,\\ \mu_3&=\alpha_3-\alpha_2\alpha_1-\tfrac{1}{3}\alpha_1^3\ ,\\ \mu_4&=\alpha_4-\tfrac{1}{2}\alpha_2\alpha_2-\alpha_3\alpha_1+\alpha_2\alpha_1^2-\alpha_1^4/4\ ,\end{aligned} \tag{A16}$$

and so on. These cumulants have somewhat different numerical factors due to the time ordering in (A13) and absence of a factorial in the expansion.

Equation (18) and the expansion (A15) is used to derive (21) for $S(Q,t)$. The expansion (A15) determines (21) directly and the coefficients μ_n. However, once we know that an expansion of the form (21) exists, the μ_n are more easily obtained in terms of the moments, M_n, by substituting (21) into (24) and determining the μ_n in terms of M_n. This leads directly to (14). The expansion (A13) is also useful for calculating expressions for the central moments of $S(Q,\omega)$ defined in (24).

APPENDIX B

In this appendix we write out the contributions B_5 and B_6 to the fifth and sixth moments of the incoherent $S_i(Q,\omega)$ listed in (30). The incoherent moments are defined in (13). They can be evaluated straightforwardly by substituting the expansion (A13) in the incoherent limit into (13). In the incoherent limit only the $\alpha_n(t)$ for even n in (A13) survive. The B_n terms come from differentiating $\alpha_4(t)$ and

$$\begin{aligned}B_5&=iQ^4[3\langle\dot{v}_Q v_Q^3\rangle+2\langle v_Q\dot{v}_Q v_Q^2\rangle+\langle v_Q^2\dot{v}_Q v_Q\rangle]\ ,\\ B_6&=-Q^4[6\langle\ddot{v}_Q v_Q^3\rangle+3\langle v_Q\ddot{v}_Q v_Q\rangle+\langle v_Q^2\ddot{v}_Q v_Q\rangle\\ &\quad+8\langle\dot{v}_Q\dot{v}_Q v_Q\rangle+4\langle\dot{v}_Q v_Q\dot{v}_Q v_Q\rangle\\ &\quad+3\langle v_Q\dot{v}_Q\dot{v}_Q v_Q\rangle]\ .\end{aligned} \tag{B1}$$

This result, in a somewhat different form, is given by Sears.[20]

For a solid in which the phonons are statistically independent and we can define a density of phonon states $g(\omega)$, the B_5 and B_6 take a simple form

$$\begin{aligned}B_5&=10A_3I_2=10\bar{a}_3\langle k_Q^2\rangle q^4\ ,\\ B_6&=15A_4I_2+10A_3^2=(15\bar{a}_4\langle k_Q^2\rangle+10\bar{a}_3^2)q^4\ .\end{aligned} \tag{B2}$$

This result (B2) also holds in any case that we can approximate $\langle v(t_1)v(t_2)v(t_3)v(t_4)\rangle$ as a product of pairs,

$$\langle v_1v_2v_3v_4\rangle\approx\langle v_1v_2\rangle\langle v_3v_4\rangle+\langle v_1v_3\rangle\langle v_2v_4\rangle+\langle v_1v_4\rangle\langle v_2v_3\rangle\ , \tag{B3}$$

where $v_1=v(t_1)$. Equation (B3) holds in the equal time limit for a Gaussian velocity distribution—i.e., in the classical limit. In normal and superfluid ^{4}He we have

found that μ_{5i} is proportional to Q^4 with the Q^2 term undetectable. Apparently the approximation (B3) does not hold well in a quantum fluid. It would be interesting to see whether it holds in a classical fluid, such as liquid argon, or in solid helium.

APPENDIX C

In this appendix we indicate the origins of the model $n(\mathbf{k})$ in (56), particularly $f(\mathbf{k})$ and $f_i(\mathbf{k})$. The single-particle momentum distribution (56) is generally related to the single-particle spectral function $A(k,\omega)$ by

$$n_k=\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}A(k,\omega)n_B(\omega)\ . \tag{C1}$$

Here n_k is normalized to $\sum_k n_k=N$, i.e., $n_k=1$ if state k is occupied, and $n(\mathbf{k})$ in (56) is $n(\mathbf{k})=n_k/(2\pi)^3 n$. In superfluid ^{4}He the single particle and density response are coupled via the condensate. The two functions have a common, sharp peak at low k. This peak is observed in the density response of superfluid ^{4}He (i.e., it is the phonon of the phonon-roton curve). The single-particle response function is

$$A(k,\omega)=2\pi z_k[\delta(\omega-ck)-\delta(\omega+ck)]+A_{\rm inc}(k,\omega) \tag{C2}$$

at low k. The first term comes from the sharp phonon (common-density–single-particle) peak, which has weight

$$z_k=\frac{n_0 Mc}{2\hbar n}\frac{1}{k} \tag{C3}$$

(Gavoret and Nozières,[47] and Griffin[48]). The sharp component is expected to dominate at $k\to 0$. Substituting (C3) and (C2) into (C1), the first term of (C2) leads directly to $f(\mathbf{k})$ in (57). In (57) we have added $e^{-k^2/2k_c^2}$ to cutoff $f(k)$ at higher k. For example, $z_k\delta(\omega-ck)$ is valid only at small k, where there is a single sharp peak only ($k\lesssim 0.4$ Å^{-1}). At higher k, the response is a sharp peak plus a broad component. Linear dispersion $\omega=ck$ is also assumed. The $f(\mathbf{k})$ in (57) certainly cannot be used beyond $k\simeq 0.7$ Å^{-1}, where the phonon region of phonon-roton curve ceases to be linear.

At higher k (i.e., certainly for $k\gtrsim 0.4$ Å^{-1}) there are broad, incoherent components to the coupled single-particle–density-response function, which contribute to $A_{\rm inc}(k,\omega)$. These will be proportional to n_0. Little is known about this part of $A_{\rm inc}(k,\omega)$, and we denote its contribution to $n(\mathbf{k})$ by

$$A_i f_i(\mathbf{k})=A_i(2\pi k_i^2)^{-3/2}e^{-k^2/2k_i^2}\ . \tag{C4}$$

A_i is a parameter, and we select $k_i=2$ Å^{-1} so $f_i(\mathbf{k})$ contributes up to high k. Finally, there is a large contribution to $A_{\rm inc}(k,\omega)$ independent of n_0, which leads to $n^*(\mathbf{k})$ describing the atoms above the condensate. We assume that $n^*(\mathbf{k})$ in superfluid ^{4}He is the same as $n(\mathbf{k})$ in normal ^{4}He, i.e., given by (51) with parameters k_0^2 and δ given in Table II. The condensate contribution $n_0\delta(\mathbf{k})$ is not captured by (C1) and must be added to (56).

APPENDIX D

In this appendix, we discuss fitting procedures. The χ^2 was defined as

$$\chi^2=\sum_{i=1}^{N_i}\left[\frac{f_i-d_i}{\sigma_i}\right]^2\Big/(N_i-N_p+1)\ ,$$

where f_i is the fitted function and d_i is the data at point i. N_i and N_p are the number of data points and free fitting parameters in f_i, respectively. In $S(Q,\omega)$, typically $N_i=75$. The χ^2 should lie between 0.8 and 1.2, approximately. A larger value signals a poor fit, a lower value signals too much error in σ_i.

In the AA we found $\chi^2=1.02$, 0.88, and 0.88 at $Q=8$ Å^{-1} and $T=2.5$ (Fig. 1) using free parameters μ_2,μ_3; μ_2,μ_3,μ_4; and μ_2,μ_3,μ_4,μ_5, respectively. This suggested including μ_5 did not improve the fit. Also μ_3 and μ_5 appeared highly correlated. Thus only μ_2,μ_3,μ_4 were retained here.

In the CA, χ^2 decreased out to and including μ_5. At $Q=8$ Å^{-1}, $T=2.5$ K (Fig. 1) $\chi^2=0.96$ in the CA including μ_5. We also tested the Q dependence of the $\mu_n(Q)$. An erratic variation of $\mu_n(Q)$ with Q suggested μ_n was not determined. We found μ_5/Q^4 versus Q was reasonably uniform and approximately independent of Q. In contrast $\mu_6(Q)$ varied erratically with Q and was not considered well determined.

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