

This expression includes both the results of Domb and those of GCS and in part answers the present question about which of the two previous developments apply to which physical system. Domb's expressions fit the Ising models but suggest corrections whose exponents are too large for the

real fluids, whereas the GCS expansions apply to fluids but seem inadequate for the magnetic Ising systems.<sup>9</sup> Application of this enlarged equation to real physical systems will however require further explicit knowledge about the structure of the lower  $G_{ijk}(x)$  functions.

<sup>1</sup>M. S. Green, M. J. Cooper, and J. M. H. Levelt Sengers, *Phys. Rev. Letters* **26**, 492 (1971).

<sup>2</sup>M. J. Cooper, in *Proceedings of Battelle Colloquium on Critical Phenomena, Geneva, 1970* (McGraw-Hill, New York, to be published).

<sup>3</sup>C. Domb, in *Critical Phenomena, Proceedings of International School of Physics Enrico Fermi* (Academic, New York, to be published); see also remarks by C. Domb given in Ref. 2.

<sup>4</sup>D. S. Gaunt and C. Domb, *J. Phys. (Paris)* **32S**, C1-344 (1971).

<sup>5</sup>D. C. Hemmer and G. Stell, *Phys. Rev. Letters* **24**, 1284 (1970), and unpublished report.

<sup>6</sup>D. S. Gaunt and G. A. Baker, *Phys. Rev. B* **1**, 1184 (1970).

<sup>7</sup>M. J. Cooper, *J. Res. Natl. Bur. Std.* **75A**, 103 (1970); also *J. Phys. (Paris)* **32S**, C1-349 (1971).

<sup>8</sup>B. W. Ninham, *J. Math. Phys.* **4**, 679 (1963), is cited in Ref. 3 for a discussion of the Darboux-type behavior in the Ising model.

<sup>9</sup>See discussion of data in Ref. 1 and also B. Wallace and H. Meyer, *Phys. Rev. A* **2**, 1610 (1970). Note for example that the Domb expression for the critical isotherm ( $\Delta T = t = 0$ ) gives  $H = aM^6 + bM^{26-1} + cM^{6+1/\beta}$ , while the fluid data suggests that  $\beta = 0.35$ ,  $\delta = 4.5$ , and the difference  $H - aM^6$  varies as  $H^p$ ,  $p = \delta + (2 \pm 0.5)$ .

## X-Ray Scattering from Liquid $^4\text{He}$ <sup>†</sup>

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The liquid structure factor of  $^4\text{He}$  has been measured in the momentum transfer range from 0.133 to 1.125  $\text{\AA}^{-1}$  and a variety of temperatures from 0.38 to 4.6 K. At the lowest temperature, the structure factor demonstrates a gentle change of slope in the momentum transfer region near 0.4  $\text{\AA}^{-1}$ . Values for the first and second moments of the pair correlation function are obtained at 1.45 and 3.30 K. We compare our results with recent neutron and x-ray work and also with recent theories.

### I. INTRODUCTION

X rays have been used as a probe to study the structure of liquid helium for the past twenty years.<sup>1-6</sup> In particular, their use allows a determination of the structure factor  $S(k)$  of the fluid. The structure factor is a measure of the correlation between positions of the atoms in the fluid. In the simplest terms, the structure factor is a measure of the diffraction produced by the sample under study compared to the diffraction that would be produced by an ideal gas. It is conventional to define the structure factor as the Fourier transform of the pair correlation function  $g(r)$  through the expression

$$S(k) = 1 + \rho \int e^{i\vec{k}\cdot\vec{r}} [g(\vec{r}) - 1] d^3r, \quad (1)$$

where  $k$  is the momentum transferred to a helium atom in a scattering event. The momentum transfer is related to the angle of scattering through the expression

$$|k| = (4\pi/\lambda) \sin \frac{1}{2}\theta, \quad (2)$$

where  $\lambda$  is the wavelength of the x ray and  $\theta$  is the angle of scattering. In the case where the positions of the atoms are uncorrelated, i. e., an ideal gas, the integral vanishes and  $S(k)$  is strictly unity for all values of  $k$ .

The intensity of x rays scattered from a typical nonideal fluid sample is given by

$$I = ANT(\sigma_e S + \sigma_i), \quad (3)$$

where  $A$  is the Thompson cross section,  $N$  the number of atoms in the sample which contribute to the scattering, and  $T$  the transmission coefficient. The quantities  $\sigma_e$  and  $\sigma_i$  are the elastic and inelastic scattering factors and  $S$  is the structure factor. In practice, one measures the structure factor as a function of  $k$  by measuring the scattering intensity as a function of scattering angle.

In the work we shall describe here we have mea-

sured the structure factor of  ${}^4\text{He}$  in the range  $0.133 \text{ \AA}^{-1} < k < 1.125 \text{ \AA}^{-1}$  at a variety of temperatures. Our current results are consistent with our previous observations<sup>5</sup> at the lowest temperatures.

## II. STRUCTURE FACTOR AT SMALL MOMENTUM TRANSFER

### A. Finite-Temperature Case

Goldstein<sup>7</sup> has shown that when the temperature is finite the structure factor is given by

$$S(k) = n_0 k_B T X_T + \sum_{n=1}^{\infty} (-1)^n k^{2n} r_G^{2n} [(2n+1)!]^{-1}, \quad (4)$$

where  $n_0$  is the number density,  $k_B$  the Boltzmann constant, and  $X_T$  the isothermal compressibility.  $r_G^{2n}$  is the moment of the pair correlation function defined by

$$r_G^{2n} = \int r^{2n} g(r) d^3r. \quad (5)$$

In general, (4) remains valid so long as  $k \rightarrow 0$  faster than  $T$ . In particular, to order  $k^6$ , Eq. (4) is just

$$S(k) = n_0 k_B T X_T - B(1) (k^2/6) + B(2) (k^4/120) - B(3) (k^6/5040) + \dots \quad (6)$$

In the limit of zero momentum transfer we immediately have

$$\lim_{k \rightarrow 0} S(k) \equiv S(0) = n_0 k_B T X_T. \quad (7)$$

Goldstein has pointed out that one expects  $r_G^2$  to change sign at some temperature which is characteristic of the system under study.

### B. Zero-Temperature Case

Feynman has suggested<sup>8,9</sup> that at absolute zero an excitation in liquid helium can be represented by a wave function of the form

$$\Psi = \phi \sum_i f(r_i), \quad (8)$$

where the sum includes all the atoms in the liquid and  $\phi$  is the ground-state wave function of the liquid. By demanding that  $\Psi$  be an eigenfunction of the total momentum, Feynman deduced that  $f(r_i) = e^{i \vec{k} \cdot \vec{r}_i}$  and hence that

$$\Psi = \phi \sum_i e^{i \vec{k} \cdot \vec{r}_i}. \quad (9)$$

He then determined an upper limit for the energy of the lowest excitation of momentum  $\hbar k$  by using the variational principle. He found that

$$E(k) \lesssim \hbar^2 k^2 / 2m S(k), \quad (10)$$

where  $S(k)$  is the liquid structure factor and  $m$  is the mass of a helium atom.

Feynman has argued that for  $k$  small enough, (9)

is the exact wave function for an excitation. These long-wavelength excitations are phonons and hence he concludes that

$$E(k) = \hbar k c = \hbar^2 k^2 / 2m S(k), \quad (11)$$

where  $c$  is the velocity of first sound in He II. Hence, Feynman predicts

$$S(k) = \hbar k / 2m c. \quad (12)$$

Expression (12) can also be derived more generally by examining the sum rules

$$S(k) = \int_0^{\infty} S(k, \omega) d\omega, \quad (13)$$

$$\frac{\hbar^2 k^2}{2m} = \int_0^{\infty} S(k, \omega) \omega d\omega, \quad (14)$$

and

$$\frac{1}{2m c^2} = \lim_{k \rightarrow 0} \int_0^{\infty} \frac{S(k, \omega)}{\omega} d\omega. \quad (15)$$

Taken together, (14) and (15) imply that<sup>10,11</sup>

$$\lim_{k \rightarrow 0} \frac{S(k)}{k} \lesssim \frac{\hbar}{2m c}, \quad (16)$$

which is equivalent to (12).

### C. Existence of a Shoulder on $S(k)$

On the basis of neutron scattering data, Jackson<sup>12</sup> and independently Miller, Pines, and Nozieres<sup>13</sup> noticed that the data available for the liquid structure factor did not seem to approach the Feynman limit. The latter authors suggested that this discrepancy could be resolved if the structure factor possessed a shoulder.

Briefly, they suggested that single quasiparticle excitations from the zero-momentum state contribute a term  $Z(k)S(\omega - \omega(k))$  to the dynamic structure factor  $S(k, \omega)$ , where  $Z(k)$  is essentially the differential cross section for single quasiparticle production. [ $Z(k)$  was measured by Henshaw and Woods<sup>14</sup> and has recently been measured to higher accuracy by Woods and Cowley.<sup>15</sup>] After considering other contributions, they suggest that this term is dominant in the long-wavelength limit. In particular from (13) they claim that

$$\lim_{k \rightarrow 0} S(k) = Z(k). \quad (17)$$

To make the entire  $S(k)$  curve compatible with the measured  $S(k)$  (above  $0.8 \text{ \AA}^{-1}$ ) and with the expected  $S(k) = Z(k)$  at very small values of  $k$ , Miller, Pines, and Nozieres suggested that there might be a shoulder on the  $S(k)$  curve near  $0.6 \text{ \AA}^{-1}$ . Since it was not known for how large a value of  $k$  one would retain  $S(k) = Z(k)$ , they could not suggest the strength of the shoulder.

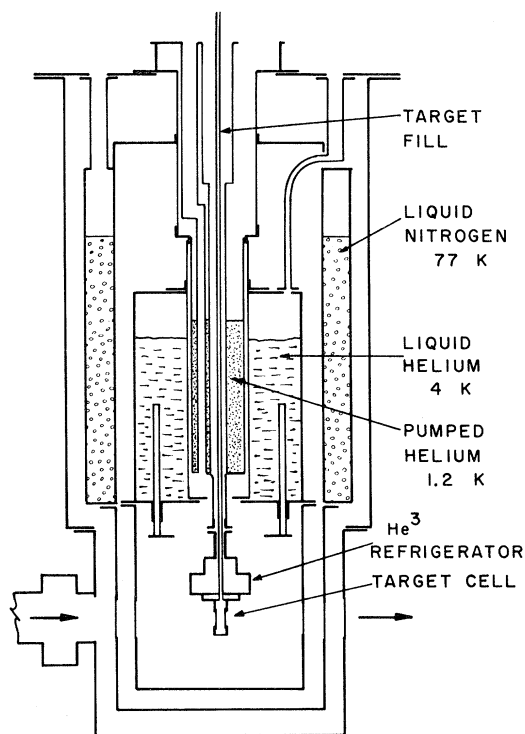


FIG. 1. Cryostat assembly. During helium and nitrogen transfers, which are done every twelve hours, the  $^3\text{He}$  refrigerator can be isolated and kept at 0.3 K. This allows filling of the 1.2 K bath without disturbing the target. The box at the left on the beam axis represents the last set of incident collimation slits. The wall between the cryostat and the detector arm is made from 0.010-in. beryllium, while the windows on the 77 and 4 K radiation shields are made from 0.001-in. aluminized Mylar.

Massey<sup>16</sup> attempted to calculate the form of the liquid structure factor by performing a variational calculation. Using the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) equation<sup>17</sup> and the Kirkwood superposition approximation,<sup>18</sup> Massey related the pair correlation function  $g(\vec{r})$  to a Jastrow-type trial function. Through (1) he was able to obtain  $S(k)$ . Massey pointed out that the weak local maximum exhibited by his structure factor near  $0.6 \text{ \AA}^{-1}$  could only be taken as suggestive. Other calculations by Lee<sup>19</sup> and Kulas<sup>20</sup> have resulted in a weaker shoulder than that given in Massey's original work.

More recently, Campbell and Feenberg<sup>21</sup> have extended Massey's work by using a paired-phonon analysis in the hypernetted chain approximation.<sup>11</sup> Their work is in reasonable agreement with our current results. Most recently, Francis, Chester, and Reatto<sup>22</sup> have obtained the liquid structure factor using a wave function originally suggested by Reatto and Chester.<sup>23</sup> Their wave function accounts for the hard core of the atoms as well as taking into

account long-wavelength modes of the fluid. This calculation results in a qualitatively correct shoulder on the structure factor in the region where the long-wavelength character begins to give way to the effects of the hard core. More theoretical work remains to be done to bring about quantitative agreement.

### III. APPARATUS

The spectrometer, cryostat, and target cells were built in our laboratory. The cryostat, modified slightly from the original designed by Safrata,<sup>24</sup> is shown in Fig. 1. The  $^3\text{He}$  refrigerator is a typical cyclic device capable of maintaining the target at temperatures near 0.3 K for several weeks at a time. Vacuum levels inside the cryostat are in the  $10^{-7}$ – $10^{-8}$  Torr range during normal operation. The target chamber is filled by condensing helium gas at about 8 liquid  $\text{cm}^3$  per hour in a thin tube<sup>25</sup> which passes directly through the  $^3\text{He}$  refrigerator. Condensation temperature is generally near 0.5 K. This design ensures that the coldest spot in the system is between the sample and the fill tube. No evidence of film flow heating was ever observed at the target. This design offers obvious advantages over the mechanical plus often used in studies of  $\text{He}^4$ .

A variety of target designs was used in this work. Details of a demountable Mylar cell<sup>26</sup> used during the early stages of this work are published else-

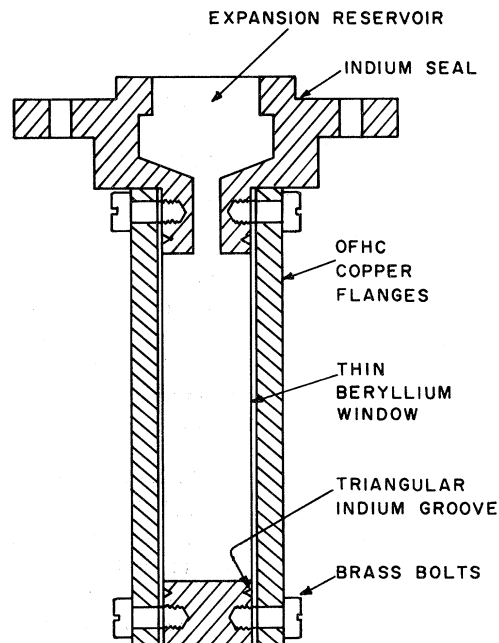


FIG. 2. Beryllium target cell. Windows 0.010 in. thick made from beryllium are sealed to two sides of this target chamber using indium.

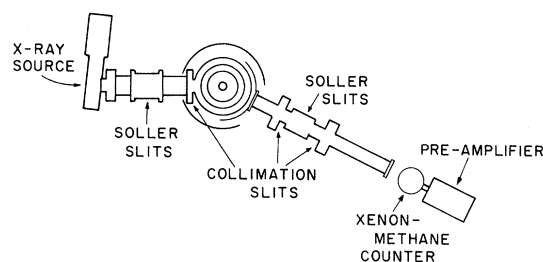


FIG. 3. Spectrometer top view (schematic). The relative position of the collimation slits can be seen. The entire spectrometer was mounted on a rigid movable experimental table.

where. Most of the data reported here were taken with the flat cell with beryllium windows shown in Fig. 2. These cells could be positioned so that their vertical axis was coincident with the rotation axis of the scattered beam detector.

A generalized top view of the spectrometer is shown in Fig. 3. A complete description of the spectrometer is given elsewhere.<sup>27</sup> Changes in scattering angle are accomplished by use of a dc stepping motor.<sup>28</sup> Single steps result in angle changes of about 0.05 deg. Data points to be presented later are separated by 10 steps or about 0.5 deg. The initial alignment and angle calibration of the spectrometer was carried out using a He-Ne laser and a diffraction grating. This calibration was checked several times during data collection by absolute measurements made on the spectrometer table surface. All of these calibration measurements agreed to within 0.05 deg for any given angle.

The x-ray beam was derived from the line source of a General Electric CA-7H (copper) x-ray tube.<sup>29</sup> Collimation was accomplished by means of a series of lead plated brass slits—each side of which could be independently positioned to better than 0.0002 in. during initial alignment.<sup>27</sup> Vertical divergence was reduced by using Soller slits<sup>29</sup> whose pitch relative to the beam axis could be adjusted to obtain maximum transmission. It should be pointed out that the x-ray beam remained in vacuum except for two gaps of  $\frac{1}{4}$  in., one between the cryostat and the detector arm and the other between the detector arm and the counter. Even though all the collimation slits were in vacuum, they could be adjusted at any time during operation. The angular resolution was measured to be about 0.55 deg by sweeping the (protected) detector (sealed xenon-methane proportional counter<sup>30</sup>) through the main beam.

Since the copper source generates a Cu K beam which is a mixture of about 88%  $K\alpha$  and 12%  $K\beta$ , a nickel filter was used in the scattered arm to reduce the  $K\beta$  contribution to 1% of the  $K\alpha$  contribution. The pulses from the detector were observed on a pulse-height analyzer, and a single-channel

analyzer (SCA) was used to eliminate all the pulses outside the Cu K peak. The combination of nickel filter and SCA resulted in a reasonable compromise between spectral purity of the beam and intensity. In normal operation the spectrometer was entirely automatic. Once the SCA was set, the analyzer was used as a scalar bank, and as data was automatically recorded at various angles, it was stored in respective analyzer channels.

The gain of the system and the room background were checked from time to time throughout the weeks of data collection, and no drifts of any significance were observed.

The temperature at the  $^3\text{He}$  refrigerator-target junction was recorded using carbon composition resistors<sup>31</sup> which were calibrated against the vapor pressure of  $^3\text{He}$  and  $^4\text{He}$ .

#### IV. COLLECTION AND ANALYSIS OF DATA

As we have seen, the intensity of x rays scattered from a typical nonideal fluid sample is given by

$$I = ATN(\sigma_e S + \sigma_i) \quad (3)$$

It is readily seen that  $N$  depends upon the scattering geometry. Tweet<sup>2</sup> long ago suggested that one way to remove this geometrical factor was simply to scatter from an ideal gas (for which  $S = 1$ ) using the same geometry as was used for the liquid scattering. In such a case we have

$$I_G = AT_G N_G (\xi_e + \xi_i) \quad (18)$$

where the  $\xi$  are the scattering factors of the ideal gas. Since the scattering volume (geometry) for a fixed angle of scattering is fixed, we can immediately obtain

$$S = \frac{IT_G \rho_G}{I_G T \rho} \left( \frac{\xi_e + \xi_i}{\sigma_e} \right) - \frac{\sigma_i}{\sigma_e} \quad (19)$$

where the  $\rho$  are number densities.

It is this expression which we have used to determine the structure factor. Neon at 77 K was used as the ideal gas.<sup>32</sup> The neon density was obtained from the Berlin virial equation<sup>33</sup> while that of liquid helium was obtained from the work of Kerr and Taylor.<sup>34</sup> The transmission coefficients were taken from published values.<sup>35</sup> Finally, the scattering factors for helium were obtained from the work of Kim and Inokuti<sup>36</sup> while those for neon were obtained from the work of Tovard *et al.*<sup>37</sup> by a fitting procedure.

To obtain the liquid structure factor, data of three kinds were required: empty target scattering, scattering from the normalizing gas (neon), and scattering from the liquid helium. A typical data run always consisted of empty target scattering taken after scattering with a sample (gas or liquid) present. In general, background counts and linear

TABLE I. Liquid structure factor of  $^4\text{He}$  determined directly from expression (19). The columns headed by a  $\sigma(\%)$  represent the expected one-standard-deviation error due to counting statistics alone and expressed as a percent. Since the window scattering is large at small angles, a very slight misalignment of the spectrometer zero-angle position from run to run can result in errors at these angles. The structure-factor values printed in parentheses may have errors two to three times as large as those listed, owing to this problem. For momentum transfer values above about  $0.3 \text{ \AA}^{-1}$  the  $T=1.45 \text{ K}$  structure factor becomes indistinguishable from the  $T=0.38 \text{ K}$  structure factor (see Figs. 4 and 7).

$k(\text{\AA}^{-1})$	$T=0.38 \text{ K}$		$T=1.45 \text{ K}$		$T=3.30 \text{ K}$		$T=4.00 \text{ K}$		$T=4.60 \text{ K}$	
	$S(k)$	$\sigma(\%)$	$S(k)$	$\sigma(\%)$	$S(k)$	$\sigma(\%)$	$S(k)$	$\sigma(\%)$	$S(k)$	$\sigma(\%)$
0.133	(0.0479)	3.28	(0.0643)	3.25	(0.200)	2.05	(0.381)	1.85	(0.449)	1.83
0.165	(0.0577)	2.42	(0.0735)	2.55	(0.199)	1.78	(0.359)	1.61	(0.423)	1.59
0.198	0.0690	2.07	0.0801	2.31	0.196	1.71	0.329	1.56	0.391	1.53
0.230	0.0764	1.95	0.0831	2.23	0.181	1.72	0.305	1.55	0.360	1.52
0.263	0.0897	1.90	0.0916	2.21	0.184	1.77	0.298	1.61	0.358	1.58
0.295	0.0960	1.83	0.0960	2.15	0.177	1.76	0.272	1.61	0.327	1.58
0.328	0.104	1.79	0.105	2.07	0.177	1.76	0.260	1.63	0.306	1.59
0.360	0.114	1.81	0.112	2.10	0.178	1.82	0.252	1.69	0.307	1.65
0.392	0.121	1.78	0.120	2.05	0.178	1.82	0.249	1.70	0.284	1.67
0.425	0.131	1.78	0.130	2.04	0.185	1.84	0.246	1.74	0.286	1.71
0.457	0.135	1.80	0.133	2.05	0.180	1.88	0.236	1.78	0.276	1.74
0.490	0.141	1.81	0.139	2.06	0.179	1.92	0.230	1.82	0.268	1.78
0.522	0.153	1.85	0.149	2.09	0.185	1.98	0.238	1.87	0.279	1.83
0.554	0.157	1.86			0.192	1.98	0.236	1.89	0.271	1.86
0.586	0.166	1.91			0.198	2.03	0.239	1.96	0.267	1.93
0.618	0.167	1.93			0.200	2.05	0.241	1.98	0.273	1.94
0.649	0.173	1.94			0.201	2.07	0.246	1.99	0.266	1.98
0.681	0.180	2.01			0.208	2.14	0.242	2.08	0.270	2.05
0.713	0.194	2.04			0.216	2.19	0.261	2.11	0.290	2.08
0.745	0.197	2.06			0.224	2.19	0.258	2.14	0.284	2.11
0.777	0.206	2.08			0.227	2.23	0.268	2.15	0.293	2.13
0.808	0.212	2.13			0.232	2.28	0.262	2.23	0.295	2.19
0.840	0.218	2.15			0.239	2.30	0.277	2.23	0.304	2.21
0.872	0.231	2.20			0.256	2.33	0.287	2.29	0.317	2.26
0.904	0.237	2.25			0.257	2.40	0.292	2.34	0.319	2.31
0.935	0.258	2.33			0.273	2.48	0.307	2.43	0.341	2.39
0.967	0.258	2.39			0.282	2.52	0.320	2.46	0.349	2.43
0.999	0.267	2.37			0.283	2.52	0.326	2.45	0.354	2.42
1.031	0.270	2.42			0.292	2.56	0.320	2.51	0.358	2.47
1.062	0.291	2.51			0.310	2.64	0.355	2.57	0.377	2.56
1.094	0.292	2.50			0.310	2.64	0.349	2.58	0.370	2.57
1.125	0.319	2.58					0.380	2.65	0.413	2.62

system gain were checked before and after every data run. Data were usually taken for about  $10^4$  counts at each angle and the sweeps of thirty or so angles were usually repeated in an attempt to improve statistics and check on reproducibility. After scattering from the sample under consideration, several empty target runs were always done. No changes in empty target scattering were observed throughout the experiment. This indicates that nothing condensed on the cryostat windows or target surface during the course of this work.

In general, the data were analyzed in the following manner. The reduced data (scattering from the fluid minus empty target scattering and room background) was inserted directly into expression (19) and  $S(k)$  determined (Table I). Various fitting expressions were then used to obtain smooth struc-

ture factor values. The smoothed values presented in Table II were produced by a weighted least-squares polynomial fit of the form

$$S(k) = \sum_{n=0}^5 a_n k^n \quad (20)$$

The choice  $n=5$  for the highest order was arbitrary. Other fitting functions (which we shall discuss in detail shortly) perhaps have more physical significance, but the results presented in Table II should serve for many purposes.

In principle, one can determine the moments of the pair correlation function  $r_G^{2n}$  by fitting expression (4) to the data. In practice our data do not extend to small enough values of  $k$  to allow a realistic determination of these moments except at the temperatures  $T=1.45$  and  $3.30 \text{ K}$ . At these temperatures,

TABLE II. Liquid structure factor of  $^4\text{He}$  as determined by a weighted least-squares polynomial fit to order  $k^5$  to the data presented in Table I. We have included  $S(0)$  as calculated from expression (7) except for the  $T = 0.38$  K case where we have taken  $S(0) = 0$  (see Fig. 7).

$k$ ( $\text{\AA}^{-1}$ )	$T=0$ $S(k)$	$T=1.45$ K $S(k)$	$T=3.30$ K $S(k)$	$T=4.00$ K $S(k)$	$T=4.60$ K $S(k)$
0.000	0	0.0540	0.205	0.370	0.622
0.050	0.0178	0.0559	0.202	0.371	0.547
0.100	0.0354	0.0611	0.197	0.360	0.484
0.150	0.0524	0.0687	0.192	0.341	0.432
0.200	0.0685	0.0778	0.187	0.319	0.388
0.250	0.0836	0.0880	0.183	0.297	0.353
0.300	0.0976	0.0989	0.180	0.276	0.326
0.350	0.111	0.110	0.178	0.259	0.304
0.400	0.123	0.121	0.178	0.246	0.289
0.450	0.134	0.132	0.180	0.237	0.278
0.500	0.145	0.143	0.184	0.234	0.271
0.550	0.155	0.154	0.189	0.234	0.269
0.600	0.166	0.165	0.196	0.237	0.269
0.650	0.176	0.175	0.204	0.244	0.273
0.700	0.187	0.187	0.213	0.252	0.279
0.750	0.198	0.199	0.223	0.262	0.287
0.800	0.210	0.211	0.234	0.272	0.297
0.850	0.223	0.224	0.246	0.282	0.309
0.900	0.237	0.237	0.259	0.293	0.322
0.950	0.252	0.252	0.272	0.305	0.337
1.000	0.268	0.268	0.286	0.319	0.353
1.050	0.285	0.284	0.301	0.338	0.371
1.100	0.301	0.302	0.317	0.363	0.389

the structure factor has small enough curvature so that reasonable fits can be obtained even for data where  $k \geq 0.133 \text{ \AA}^{-1}$ . We shall consider these two cases and the  $T = 0.38$  K case in more detail following some comments concerning errors.

### V. ERRORS

Experiments of this nature are often prone to a variety of systematic errors. The transmission coefficient of neon is reasonably well known, but that for helium<sup>35</sup> is uncertain. We expect that this introduces a 2% systematic error into our results. We have taken the scattering factors for neon and helium as exact. Owing to its finite thickness, our sample transmitted only about 92% of the beam incident upon it when filled with liquid helium. In the case of a sample fully bathed in radiation of the size used here, one expects<sup>38,39</sup> that about 3% of our observed intensity was a result of multiple scattering. Our sample cell was only partially illuminated by radiation, however, and hence we have made a 1% angle-independent multiple-scattering correction. We expect that multiple-scattering effects may introduce as much as a 2% error. Our helium was pure, being filtered twice through liquid-nitrogen-temperature charcoal filters before it entered the cryostat. The neon<sup>40</sup> was also charcoal trapped. We expect that our total systematic error is 3–4%.

Random errors were also encountered. The

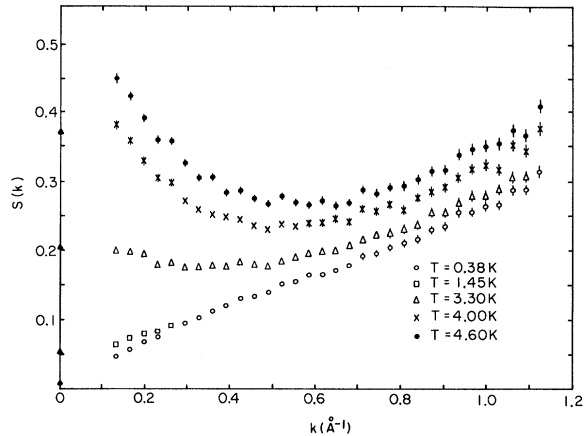


FIG. 4. Liquid structure factor at various temperatures. The closed triangle represents  $S(0)$  as calculated from expression (7).

x-ray beam was observed to be stable to about 1% from run to run. Counting statistics were generally between 1 and 2% for both the gas and liquid data. We expect therefore, that our total error does not exceed about 5–6%. The fitted curves remove most of the effects of both counting statistics and beam intensity fluctuations in any given run. We expect that the smoothed curves are accurate to 4–5%.

### VI. RESULTS AND DISCUSSION

Some of our results are shown in the figures. Error bars represent plus and minus one standard deviation due to counting statistics alone. In cases where no error bars are shown for this work, it

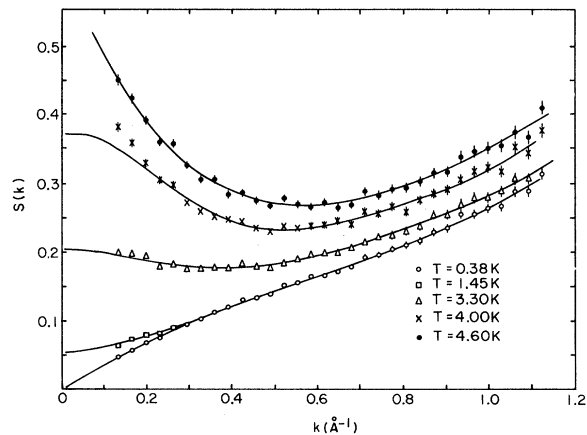


FIG. 5. Curves shown here are a result of a weighted least-squares polynomial fit to order  $k^5$  of the data presented in Fig. 4 and Table I. In each case we have included  $S(0)$  as calculated from expression (7) in our determination of the fit. In the case of the data for  $T = 0.38$  K we have set  $S(0) = 0$ .

TABLE III. Moments of the pair correlation function. The columns contain the moments of the correlation function we have obtained by fitting Eq. (4) to our data. The error bars represent a one-standard-deviation confidence level for the fit and exclude effects of systematic errors.

	$T=1.45$ K	$T=3.30$ K
$r_G^2$	$-3.65 \pm 0.14 \text{ \AA}^2$	$2.59 \pm 0.20 \text{ \AA}^2$
$r_G^4$	$-159 \pm 20 \text{ \AA}^4$	$237 \pm 30 \text{ \AA}^4$

is because they are the same size as or smaller than the plotted symbol.

In Fig. 4 we show the structure factor of  $^4\text{He}$  at various temperatures at saturated vapor pressure. In Fig. 5 we show the curves resulting from the least-squares polynomial fit [Eq. (20)] to the data of Fig. 4. Several features of the data are worthy of comment. In each case our data are reasonably consistent with the value of  $S(0)$  obtained from expression (7). The data also clearly indicate that the curvature of  $S(k)$  in the small- $k$  regime undergoes a change of sign<sup>7</sup> somewhere between 1.45 and 3.30 K. Experiments are under way at present in an attempt to determine the temperature at which this curvature appears to vanish.

As we have already indicated, one can in principle determine the moments of the pair correlation function from a fit of expression (4) to the data. We have carried out such a fitting procedure and found values (Table III) for these moments at temperatures of  $T=1.45$  and  $3.30$  K. At these temperatures, the structure factor has small curvature at small  $k$  and hence data obtained for  $k \geq 0.133 \text{ \AA}^{-1}$  can be expected to yield reasonable results. Figure 6

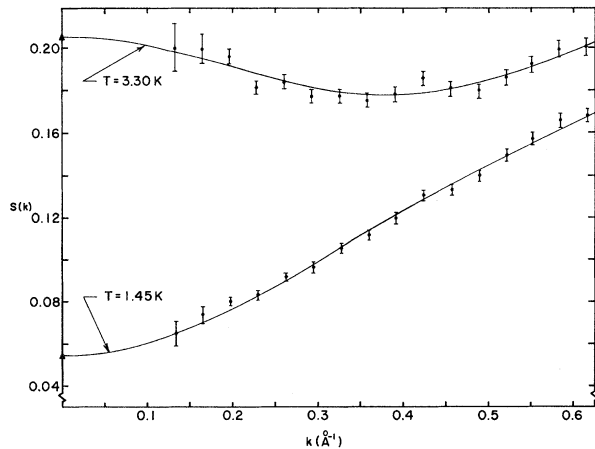


FIG. 6. Liquid structure factor at  $T=1.45$  and  $3.30$  K. The points are the experimental structure factors from Table I. The smooth curves result from fitting Eq. (4) to the data.

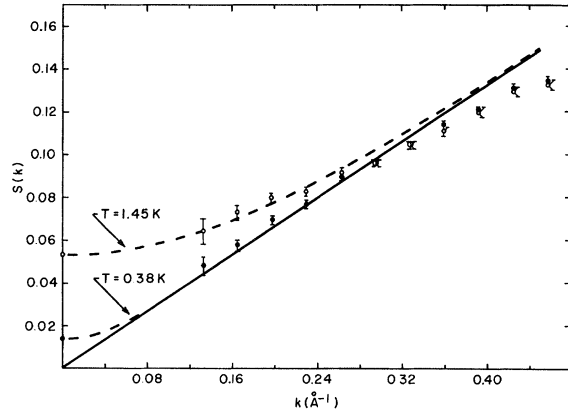


FIG. 7. Liquid structure factor at  $T=0.38$  and  $1.45$  K. The dashed curves are obtained from Eq. (21) at the indicated temperatures. As is evident from the figure, our data at  $0.38$  K do not extend to small enough angles to exhibit the characteristic curvature imposed by the hyperbolic cotangent. Rather, our results at  $0.38$  K confirm the linearity expected from Eq. (12). As can be seen from the figure, for values of momentum transfer below  $0.2 \text{ \AA}^{-1}$  the measured structure factor at  $0.38$  K lies slightly above the line given by the Feynman relation. Although at the edge of statistics, the presence of this slight effect in earlier results (Ref. 5) suggests that further experiments of higher accuracy at small values of and low values of  $T$  should be carried out. These experiments are beyond the planning stage.

shows our data at these two temperatures along with curves drawn from the fits given in Table III. The values one obtains for the coefficients of the fit depend to some extent on the number of data points one chooses to fit. The coefficients given in Table III result from a fit to the data for  $k \leq 0.65 \text{ \AA}^{-1}$ . At temperatures where the curvature in the small- $k$  structure factor is large, our data do not extend to small enough angles to give a meaningful fit.

Figure 7 displays our data at  $0.38$  and  $1.45$  along with the function

$$S(k) = (\hbar k / 2mc) \coth(\hbar kc / 2k_B T), \quad (21)$$

evaluated at each temperature. Equation (21) was obtained by Feynman and Cohen<sup>9</sup> as an intuitive extension of the zero-temperature result of Feynman,  $S(k) = \hbar k / 2mc_1$ . At the smallest values of the momentum transfer, our results are in accord with Eq. (21). At these small values of  $k$  one can expand the hyperbolic cotangent to obtain

$$S(k) = k_B T / mc^2 + A(k^2/6) + B(k^4/120) + \theta(k^6), \quad (22)$$

where

$$A = \frac{\hbar^2}{2mk_B T}, \quad B = -\frac{\hbar^4 c^2}{6mk_B^3 T^3}.$$

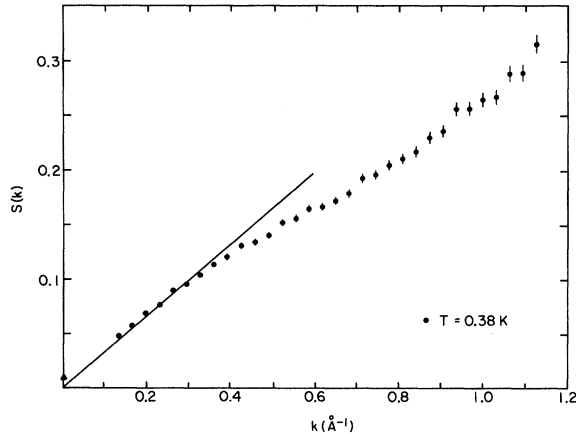


FIG. 8. Liquid structure factor at low temperature. The straight line is the structure factor one expects from the Bjel-Feynman dispersion relation. Again, the solid triangle represents  $S(0)$  as calculated from expression (7).

The quantity  $\hbar^2/2mk_B T$  is the square of the mean thermal de Broglie wavelength  $\lambda_T^2$  of a helium atom and has the value  $4.14 \text{ \AA}^2$  at  $T = 1.45 \text{ K}$ . We have fit our data at the smallest angles to Eq. (22) and have found

$$A = 3.75 \pm 0.67, \quad B = 160 \pm 390, \quad T = 1.45 \text{ K}$$

in reasonable agreement with the expected coefficients.

As mentioned previously, our data at the lowest temperatures ( $0.38 \text{ K}$ ) does not extend to small enough angles for the curvature imposed by the hyperbolic cotangent to be evident, but rather, we confirm within experimental error the linear relation

$$S(k) = \hbar k / 2mc \quad (23)$$

Figure 7 also exhibits this behavior. As is shown in the figure, we cannot unambiguously distinguish our data at  $T = 0.38 \text{ K}$  from that we might expect at absolute zero. The present results are in excellent agreement with our previous work.<sup>5,27</sup> For momentum transfer values beyond about  $0.4 \text{ \AA}^{-1}$ , our data fall below the line given by Eq. (23) (see also Fig. 8). It is conceivable that some second-order effect or systematic error is responsible for this gentle change in slope. We believe the change to be real, however, as it persists through changes in target size, shape, and construction material.

Several authors have suggested the manner in which the zero-temperature structure factor  $S(k)$  might deviate from the Feynman result for values of  $k$  which are not vanishingly small. In the analysis and discussion which follows we have made the assumption that our data at  $T = 0.38 \text{ K}$  may be taken as representative of what one might expect for the

structure factor at absolute zero. In view of Fig. 7 we feel that this assumption is justified.

We have found that three parameter expansions for  $S(k)$  of the form

$$S(k) = \frac{\hbar k}{2mc} \left[ 1 + B(1) \left( \frac{\hbar k}{mc} \right)^n + B(2) \left( \frac{\hbar k}{mc} \right)^{2n} + B(3) \left( \frac{\hbar k}{mc} \right)^{3n} \right] \quad (24)$$

provide quite reasonable fits to the data over the entire range  $k \leq 1.125 \text{ \AA}^{-1}$ . For weighted fits,  $n = 1$  provides a better fit, while for fits with each point weighted equally,  $n = 2$  is better. We might point out again here that the parameters one obtains depend on how much data he chooses to fit.

When restricted to three parameters, the expansion suggested by Woo and others

$$S(k) = \frac{\hbar k}{2mc} \left[ 1 + B(1) \left( \frac{\hbar k}{mc} \right)^2 + B(2) \left( \frac{\hbar k}{mc} \right)^3 + \dots \right], \quad (25)$$

yields a slightly worse fit than Eq. (24) (for either value of  $n$  and for both weighted or unweighted fits). Table IV contains a summary of some of our fits over the full range  $k \leq 1.125 \text{ \AA}^{-1}$ .

In view of the errors involved in our experiment we do not feel that any particular fit is to be unambiguously preferred over any of the others. The data suggest, however, that if the linear term is present, it has a rather small coefficient. [See the Appendix for a discussion of the relationship of our  $S(k)$  to the excitation spectrum  $E(k)$  as measured by Woods and Cowley.]

In Fig. 9 we compare our data with the work of Gordon *et al.*<sup>1</sup> and the recent work of Achter and Meyer.<sup>6</sup> One point should be emphasized here. Although Gordon *et al.*<sup>1</sup> used helium gas for normalization, the problem of its structure did not directly influence their determination of  $S(k)$ . This is

TABLE IV. Coefficients of the three-parameter expansions Eqs. (24) and (25). The  $B(i)$  values are the coefficients of the expansions mentioned in the text. In the weighted fits we have weighted each value of  $S(k)$  by  $\pm$  one standard deviation as determined by the observed intensity. In the case of equal weighting we have assigned equal error to each  $S(k)$  value. The column headed by  $\Delta$  contains the sum-square deviation of each fit relative to the best fit in the group.

Equation	$B(1)$	$B(2)$	$B(3)$	$\Delta$
Weighted fits				
(24), $n=1$	$0.25 \pm 0.08$	$-2.75 \pm 0.30$	$2.88 \pm 0.26$	1
(24), $n=2$	$-1.42 \pm 0.09$	$2.90 \pm 0.46$	$-1.55 \pm 0.57$	1.34
(25)	$-1.59 \pm 0.27$	$1.27 \pm 0.88$	$0.66 \pm 0.72$	1.41
Equal weight fits				
(24), $n=1$	$0.086 \pm 0.119$	$-2.11 \pm 0.43$	$2.30 \pm 0.37$	1.02
(24), $n=2$	$-1.47 \pm 0.11$	$3.13 \pm 0.56$	$-1.85 \pm 0.69$	1
(25)	$-1.92 \pm 0.32$	$2.42 \pm 1.07$	$-0.31 \pm 0.88$	1.03



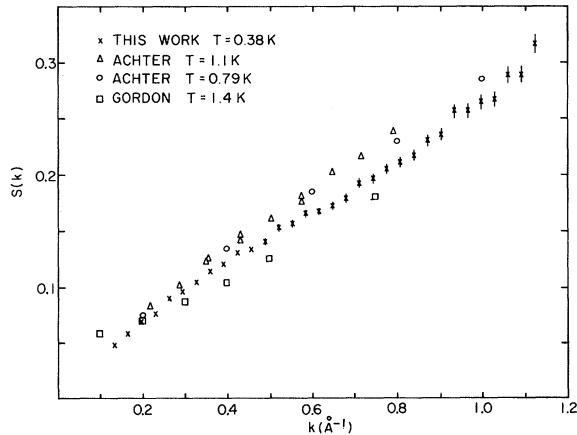


FIG. 9. Comparison of this with other work. The disagreement with the work of Achter and Meyer in the region around  $k=0.7 \text{ \AA}^{-1}$  is not understood.

because they tied their gas data to theory at a scattering angle of  $20^\circ$  and used the theoretically adjusted data rather than the real gas scattering to normalize their data at smaller angles. In this manner the severe departure from unity of the structure factor of helium gas at small momentum transfer and low temperatures<sup>5,6,27</sup> did not affect their results as much as it might have. A detailed study of the structure of helium gas has been completed at a variety of pressures and several temperatures near 5 K and will be published separately.

The agreement between our present work at the lowest temperatures and the recent theoretical work by Campbell and Feenberg<sup>21</sup> can be seen in Fig. 10. The Bijl-Feynman linearity at the smallest angles was an input to, rather than a result of, their theory. The work of Francis, Chester, and Reatto<sup>22</sup> is also shown in the figure. Their work predicts a change of slope in the structure factor, but the position and shape disagree somewhat with the present measurements.

In the final figure, Fig. 11, we show how our structure factor  $S(k)$  compares with the strength of the single-particle excitations  $Z(k)$  as recently measured by Woods and Cowley.<sup>15</sup> We see that at small angles they are essentially equal.

#### VII. EXTENSIONS OF THIS WORK

There are several obvious extensions to the work we have reported here. At the present time we are essentially repeating this work to study the small angle scattering properties of  $^3\text{He}$  as a function of temperature. It will also be a simple matter to study mixtures.

It has been suggested<sup>41</sup> that x-ray scattering techniques might be useful in observing Fermi surface effects in  $^3\text{He}$  at very low temperatures. Below the degeneracy temperature, the momentum

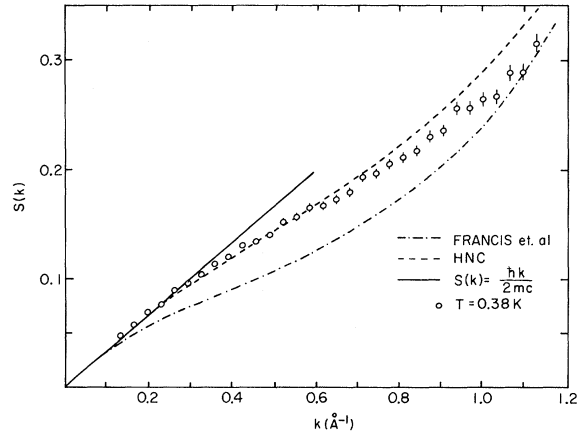


FIG. 10. Dots represent our structure factor at the lowest temperature. The solid line is the Bijl-Feynman prediction while the dashed curve is the structure factor obtained by Campbell and Feenberg (Ref. 21). The dash-dot curve represents the work of Francis *et al.* (Ref. 22).

distribution of an ideal Fermi system has a discontinuity at the Fermi surface. Luttinger<sup>42</sup> has shown that this may be true in the interacting system as well. Kohn<sup>43</sup> long ago proved that when such a discontinuity is present the response of the system will have a singularity. In the case of  $^3\text{He}$ , the liquid structure factor should have a discontinuous second derivative when the momentum transfer is equal to twice  $k_F$  (at least for the ideal case). Unfortunately, to our knowledge, no calculation of the magnitude of this jump in the case of an interacting system of  $^3\text{He}$  atoms exists.

Our spectrometer is entirely automatic and hence, it is possible to examine portions of the structure factor with a resolution of better than  $0.1^\circ$  and with

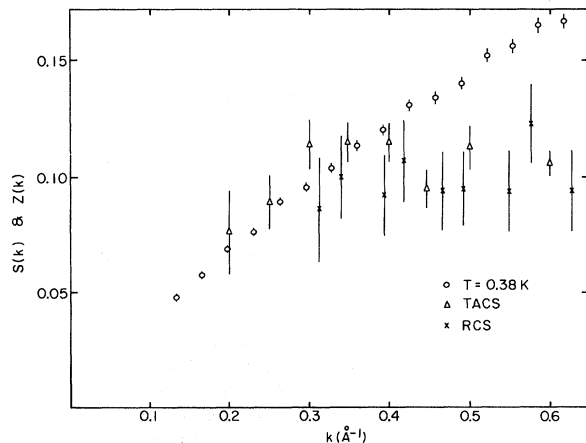


FIG. 11. Comparison of our small-angle  $S(k)$  with  $Z(k)$  as measured by Woods and Cowley (Ref. 15). TACS and RCS refer to the spectrometers used in the work of Ref. 15.

counting statistics exceeding  $\frac{1}{2}\%$  in an experiment of reasonable duration. In view of this we have constructed a dilution refrigerator and are attempting to determine  $k_F$  in  ${}^3\text{He}$ .

Since the Fermi momentum depends on the density through the relation

$$k_F = (3\pi^2\rho/m)^{1/3}, \quad (26)$$

it might be possible to dilute a sample of pure  ${}^3\text{He}$  by adding  ${}^4\text{He}$  and hence perhaps measure the dependence of  $k_F$  on the effective  ${}^3\text{He}$  density.

#### ACKNOWLEDGMENTS

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#### APPENDIX: RELATIONSHIP OF $S(k)$ TO $E(k)$

Ever since the work of Feynman demonstrated that the elementary excitation spectrum and the structure factor were closely related through

$$E(k) = \hbar^2 k^2 / 2mS(k), \quad (A1)$$

the true connection between  $E(k)$  and  $S(k)$  has been a subject of extensive study. It is the purpose of this Appendix to examine our structure factor measurements in relation to recent neutron-scattering and specific-heat results. To facilitate this comparison we shall adopt the expansion

$$S(k) = \frac{\hbar k}{2mc} \left[ 1.0 - 1.42 \left( \frac{\hbar k}{mc} \right)^2 + \dots \right], \quad (A2)$$

as representative of our results.

It is generally assumed that the dispersion relation at small  $k$  for He II has the form

$$E(k) = \hbar kc \left[ 1 - \gamma (\hbar k)^2 + \dots \right]. \quad (A3)$$

Usually,  $\gamma$  is taken to be positive,  $\gamma > 0$ . Maris and Massey<sup>44</sup> have recently suggested that  $\gamma$  may be negative. They argue that the theory of the acoustic

attenuation and the sound velocity dispersion are in better agreement with experiment under such an assumption. Phillips<sup>45</sup> and co-workers claim to have determined  $\gamma$  in a careful specific-heat measurement and found it to have a value of  $\gamma = -4.1 \times 10^{37} \text{ cm}^{-2} \text{ g}^{-2} \text{ sec}^2$  at low temperature and saturated vapor pressure.

Using our data on  $S(k)$  as represented by (A2) and Eq. (A1), we find that

$$\hbar kc \left[ 1 - \gamma_x (\hbar k)^2 \right] = \hbar k / 2mS(k) \quad (A4)$$

implies a value  $\gamma_x = -5.7 \pm 0.3 \times 10^{37} \text{ cm}^{-2} \text{ g}^{-2} \text{ sec}^2$ . As usual, the error bars result from a one-standard-deviation confidence level in the determination of the fit parameters.<sup>46</sup> Although this result is interesting, it is by no means clear that Eq. (A1) is the best relationship between  $E(k)$  and  $S(k)$  and hence  $\gamma_x$  must be viewed with caution.

In this regard, Lai, Sim, and Woo<sup>47</sup> have found

$$E(k) = \frac{\hbar^2 k^2}{2mS(k)} \left[ 1.0 - 1.46 \left( \frac{\hbar k}{mc} \right)^2 + \dots \right]. \quad (A5)$$

This result combined<sup>48</sup> with the expansions

$$E(k) = \hbar kc \left[ 1 + \omega_2 (\hbar k/mc)^2 + \dots \right], \quad (A6)$$

$$S(k) = (\hbar k / 2mc) \left[ 1 + z_2 (\hbar k/mc)^2 + \dots \right], \quad (A7)$$

which result<sup>49</sup> from sum rule arguments suggest that

$$\omega_2 = -1.46 - z_2. \quad (A8)$$

Using Eq. (A2) we find  $\omega_2 = -0.04 \pm 0.09$ , which results<sup>50</sup> in

$$\gamma = 1.6 \pm 3.6 \times 10^{36} \text{ cm}^{-2} \text{ g}^{-2} \text{ sec}^2.$$

This value is consistent with the result<sup>15</sup>  $\gamma = 0 \pm 2 \times 10^{36} \text{ cm}^{-2} \text{ g}^{-2} \text{ sec}^2$  as recently reported by Woods.

In view of the errors of our experiment and uncertainties which arise when fitting our data to the various structure factor expansions (and, in fact, the question of the validity of the expansions themselves for large  $k$ ), we view the results of this Appendix as suggestive rather than definitive.

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- <sup>30</sup>Reuter Stokes Model RSG-61 available from the Reuter Stokes Corporation, Cleveland, Ohio.
- <sup>31</sup>Speer grade 1002,  $\frac{1}{2}$  W, 220  $\Omega$  nominal, available from the Speer Carbon Corporation.
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