Measurement of the kinetic energy in ^{4}He through the superfluid transition by very high-energy neutron scattering

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Abstract. Measurements of the kinetic energy of atoms in liquid ⁴He have been made using neutron Compton scattering at wavevector transfers between 90 and 150 Å⁻¹. The kinetic energy shows a marked decrease from 16.2 K at a temperature of 2.5 K, to 13.9 K at 1.3 K. Three different measurements were made using energy analysers with different resolutions. Good agreement was obtained between the measurements using different resolutions and with previous measurements and calculations of the kinetic energies. For the best-resolution data, a line shape analysis was made in order to derive the non-Gaussian components of the momentum distribution. Values of kinetic energies are consistent with a Bose condensate fraction of ∼10%. The measurements show that eV neutron scattering provides an accurate method of measuring kinetic energies in quantum fluids.

1. Introduction

It is generally accepted that the occurrence of superfluidity in ⁴He is associated with the presence of a Bose condensate, in which a macroscopic number of particles occupy a single momentum state. It was first suggested by Hohenberg and Platzmann [1] that the Bose condensate in ⁴He could be directly observed by high-energy neutron scattering, a technique which is known as 'deep inelastic neutron scattering (DINS)' or 'neutron Compton scattering' (NCS). The former name is by analogy with the older technique of deep inelastic scattering of electrons from nuclei and the latter after Compton scattering, which is used to measure atomic momenta by the scattering of high-energy photons [2].

All three techniques rely upon the fact that when the energy and momentum transferred in the scattering process are sufficiently large, the impulse approximation (IA) is accurate. In the IA [3–6], scattering occurs from a single atom with conservation of kinetic energy and momentum. Thus the energy change ω of the neutron must be the difference between the initial and final atomic kinetic energies. If the momentum of the neutron changes by $\boldsymbol{q}$ and the initial atomic momentum is $\boldsymbol{p}$ we obtain

$$\omega = \frac{(\boldsymbol{p}+\boldsymbol{q})^2}{2M} - \frac{p^2}{2M} \tag{1}$$

where M is the atomic mass. Rearrangement of this equation gives

$$\boldsymbol{p} \cdot \hat{\boldsymbol{q}} = y = \frac{M}{q}\left(\omega - \frac{q^2}{2M}\right) \tag{2}$$

where $\hat{q}$ is the unit vector along q. Thus from a measurement of the change in energy and momentum of the neutron, the component of atomic momentum along the direction of q, which is usually denoted by y^2, can be determined. By measuring a large number of scattering events the probability distribution, $J(y)$, of atomic momentum components along the direction of q can be measured. In an isotropic system such as liquid ^{4}He, $J(y)$ is related to the momentum distribution $n(p)$ via the equation [3]

$$J(y) = 2\pi \int_{|y|}^{\infty} pn(p)\,\mathrm{d}p. \tag{3}$$

Since the suggestion of Hohenberg and Platzmann, many attempts have been made to directly observe the Bose condensate in ^{4}He using NCS. Unfortunately, as Hohenberg and Platzmann originally noted, interpretation of experimental data is complicated by the occurrence of 'final-state effects' (FSEs), which lead to a broadening of the observed $J(y)$. The physical origin of FSEs is the localization of the struck atom by surrounding atoms, after the collision with the neutron. This localization makes the momentum of the atom in the final state uncertain and hence the momentum of the atom in the initial state is also uncertain, since this is inferred from the measurement of the neutron momentum change and conservation of momentum. Corrections for these effects are rather difficult to calculate and, since FSEs are reduced as q increases, over the past three decades there has been a continuing effort to increase the energy and momentum transfers in DINS measurements on liquid ^{4}He.

The earliest attempts to measure $J(y)$ in liquid helium were made at reactor sources [7] at momentum transfers up to 15 Å^{-1}. At these relatively low momentum transfers substantial deviations from the IA are observed and this makes the interpretation of data uncertain. The range of momentum transfers available for NCS measurements has been substantially increased by the development of advanced accelerator based neutron sources, which have much more intensity than reactors at neutron energies in excess of 1 eV. The current state of the art measurements were performed by Sokol and coworkers on the Phoenix chopper spectrometer at the 'Intense Pulsed Neutron Source' (IPNS), Argonne National Laboratory, USA [8, 9], at momentum transfers of 25 Å^{-1}. However, even at these relatively large momentum transfers, final-state broadening of the postulated $\delta(y)$ condensate component in $J(y)$ is still significant and a full many-body calculation [10] is necessary to take this into account. There is a considerable literature on the form and size of FSEs in ^{4}He. The calculation of FSEs is a rather difficult many-body problem and the form of FSEs in NCS measurements is a problem of considerable interest, which has been used as a test bed for advanced many-body techniques [10–12].

In this paper we present NCS measurements on ^{4}He performed using electron volt neutrons, with momentum transfers ranging between 90 and 150 Å^{-1} and energy transfers 4–11 eV. At such large values of energy and momentum transfers, FSEs in liquid helium are essentially negligible and the rather complex data corrections required at lower energies are not necessary. There was some indication from early measurements on ^{4}He using a resonance filter spectrometer [13] that the difference between the superfluid and normal phases was larger than expected, with a condensate fraction of 30% representing the best fit to the data. One of the aims of this study is to determine whether measurements at very high momentum transfers do give significantly different results to those at lower momentum transfer, as would be the case if final-state corrections were inaccurate. Another important aim is to provide a careful check of the procedures which are used on eVS to determine atomic kinetic energies. This was done by performing three different measurements using different instrument geometries and energy resolutions and using two quite different data

analysis procedures. The best-resolution data was obtained using a prototype analyser cooling arrangement and another aim was to evaluate the improvements in data which could be obtained from improved instrument resolution at eV energies.

The experimental details are presented in section 2. In section 3 we present the results of the three measurements with different resolutions and analysis procedures. In section 4 we discuss these results and draw conclusions.

2. Experimental details

The electron volt spectrometer (eVS) has been developed specifically to take advantage of the high flux of neutron energies in excess of 1 eV which are available on the ISIS source at the Rutherford Appleton Laboratory. On eVS momentum transfers in excess of 150 Å^{-1} are accessible and NCS measurements can be made on both weakly bound systems such as quantum fluids [14–19] and more strongly bound systems, and particularly protons and deuterons in a wide variety of hosts [20–26]. Although the resolution of eVS in atomic momentum space is at present significantly worse than in the best chopper measurements, the advantage of eVS for these studies is that FSEs are essentially negligible and the count rate is significantly higher than in previous measurements.

A nuclear resonance filter difference technique [21] is used to analyse the energy of the scattered neutron. This technique utilizes foils of either gold or uranium, which absorb neutrons strongly over a narrow range of energies, centred at neutron absorption resonances. Two measurements are taken: one with the foil between sample and detector and one with the foil removed. The difference between these two measurements gives the number of neutrons captured by the foil and defines the final neutron energy with a line shape determined by the transmission of the foil. Standard time of flight techniques then yield the q and ω in the scattering process. The uranium foil provides much better resolution than the gold foil, but the gold foil provides a count rate higher by a factor ∼10 than uranium. Thus if one is mainly interested in the variation of kinetic energy with thermodynamic constraints such as temperature [15, 19], pressure [16, 17] or composition [18] then the gold foil has considerable advantages. One of the primary aims of this study was to test the accuracy with which kinetic energies can be derived, with a gold foil, when the intrinsic width of the momentum distribution is significantly less than the resolution width.

We performed three sets of measurements with different resolutions.

(1) *A measurement using a gold foil analyser covering six detectors with scattering angles between 99° and 113°.* The energy resolution of the gold foil has a Lorentzian line shape centred at 4.908 eV, with a half width at half maximum of 130 meV.

(2) *A measurement using a uranium foil analyser at room temperature, covering 16 detectors with scattering angles between 130° and 152°.* The intrinsic Lorentzian line shape of the resonance is dominated by Doppler broadening and is Gaussian, centred at 6.771 eV with a standard deviation of 63 meV.

(3) *A measurement using a uranium foil analyser cooled to 77 K in a closed cycle refrigerator, covering detectors with scattering angles between 142° and 153°.* Cooling the foil reduces the Doppler broadening of the uranium resonance line shape, but this is still well approximated by a Gaussian line shape, with a standard deviation of 49 meV.

The first two sets of measurements were performed on the same sample at five temperatures: 1.3, 1.5, 1.7, 1.9 and 2.5 K. The total counting time at each temperature was ∼20 hours. The third measurement was made in a different experiment at two temperatures, 1.5 K and 2.5 K, with a counting time of ∼80 hours at each temperature. A longer counting time

was required in the latter experiment since only eight detectors could be covered by the foil and, due to constraints of experimental geometry, it was necessary to increase the final flight path from 0.5 metres to 0.8 metres.

The resolution in momentum space of eVS is determined by uncertainties in the incident and final flight paths, the scattering angle, the measurement of the neutron time of flight and the width of the resonance absorption line in the analyser filter. These can be determined by a standard procedure, using a lead calibration sample and used to calculate the spectrometer resolution in y space [27]. The basic assumptions in the resolution calibration are that

(a) the momentum distribution of Pb at room temperature is well described by a Debye model (since the Debye temperature of Pb is 88 K, the kinetic energy of Pb atoms at room temperature differs by only 0.1% from the classical value of $3kT/2$ and even if the density of states is not Debye-like the assumed momentum distribution should still be highly accurate) and

(b) all resolution components, other than that due to the absorption resonance line shape for Au, have a Gaussian line shape. This approximation is least accurate for the contribution from the shape of the time pulse in the moderator. This introduces an asymmetry in the line shape, which can be observed in figures 1 and 2 and is currently neglected in the fitting procedures. The momentum and energy transfers and the widths of the instrument resolution function, obtained from the calibration procedure, are summarized in table 1 for some representative scattering angles and for the three different analyser foils.

Table 1. The energy and momentum transfers are listed for different foils and scattering angles. Δy_E is the contribution of the energy resolution to the spectrometer resolution in momentum (y) space. For the gold foil the value given for Δy_E is the HWHM of the Lorentzian line shape, whereas for the uranium foil the standard deviation of the Gaussian resolution function is given. Δy_I is the standard deviation of the Gaussian resolution due to all other instrument uncertainties.

| Scattering Angle (°)	Resonance foil	Resonance energy E_l (eV)	Resonance width ΔE_l (eV)	Momentum transfer q (Å^{-1})	Energy transfer ω (eV)	Δy_E (Å^{-1})	Δy_I (Å^{-1})
106	Au	4.908	0.128	94	4.58	1.86	0.71
123	U	6.671	0.063	125	8.15	0.79	0.72
137	U	6.671	0.063	136	9.64	0.75	0.75
152	U	6.671	0.063	152	10.96	0.73	0.65
148	cooled U	6.671	0.045	143	10.59	0.57	0.67

3. Results and analysis

The data from different detectors can be transformed to y space [21], where the spectra from individual detectors can be summed to improve statistical accuracy. Figures 1(a) and 1(b) show the composite spectra obtained by this procedure with the gold foil analyser at 1.4 and 2.5 K. The difference in the spectra, although small, is statistically significant and was analysed by fitting a Gaussian convolved with the calibrated instrument resolution function, i.e. we assumed that $J(y)$ has the form

$$J(y) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(\frac{-y^2}{2\sigma^2}\right). \tag{4}$$
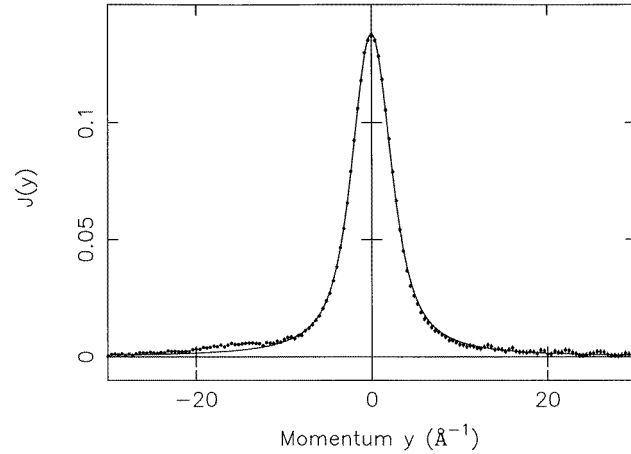
(a)
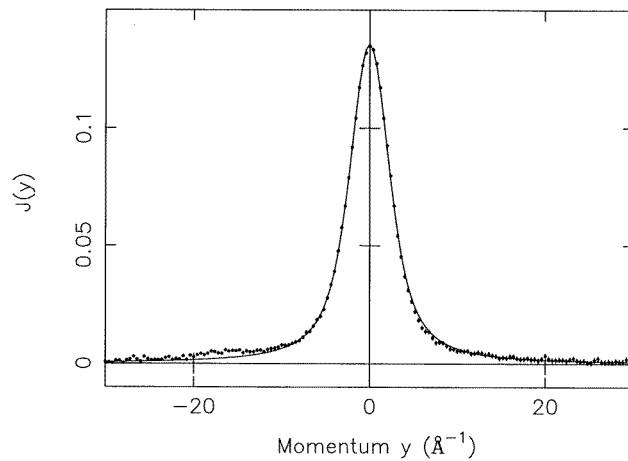
(b)

Figure 1. The sum of data from six detectors, using the Au analyser foil at room temperature. The solid line is a fit of a Gaussian, convolved with the instrument resolution function, as described in the text. (a) 1.4 K, (b) 2.5 K.

The fit is shown as the solid line in figure 1. Since the instrument resolution function varies with angle, a Gaussian, convolved with the appropriate resolution function, was fitted to the data in each of the detectors. An identical procedure was followed to analyse the data collected with the room-temperature uranium foil analyser. Corresponding plots with fits are shown in figures 2(a) and 2(b). We note that some asymmetry at low y can be observed in all four data sets, due to the asymmetry in the time structure of the neutron pulse shape, which is not taken into account in the fitting procedure.

The values of σ obtained from the fitting procedure are given in table 2. In an isotropic system, such as liquid ^{4}He, the atomic kinetic energy κ is related to $J(y)$, via the equation,

$$\kappa = \frac{3}{2M} \int y^2 J(y) \, dy = \frac{3\sigma^2}{2M}. \tag{5}$$
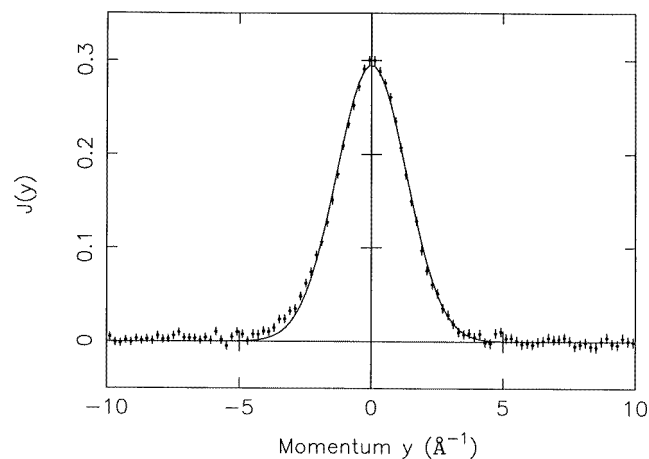
(a)
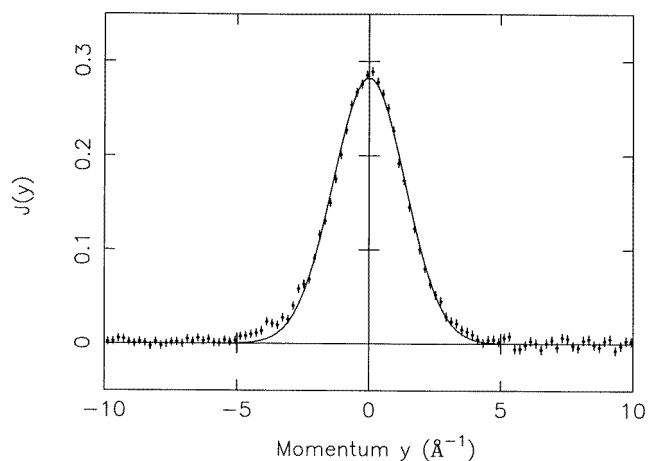
(b)

Figure 2. The sum of data from 16 detectors, using the room-temperature uranium foil analyser. The solid line is a fit of a Gaussian, convolved with the instrument resolution function, as described in the text. (a) 1.4 K, (b) 2.5 K.

The values of kinetic energy calculated from this equation are also listed in table 2. If we make the rather oversimplified assumption made in previous eV neutron studies of ^{4}He [13], that the reduction in kinetic energy is entirely due to the development of a condensate and that the average kinetic energy of the 'uncondensed' atoms is the same as in the normal phase, we obtain the values for the condensate fraction given in column 6.

The simple procedure of fitting a single Gaussian to the distribution for Au and room-temperature U data was chosen because the current instrument resolution is too poor to make any detailed lineshape analysis worthwhile. The analysis of previous measurements at lower momentum transfers [9] consisted of fitting the sum of two Gaussians to the data and the data from these previous measurements were parametrized in terms of the widths and relative weights of the two components. In order to test the accuracy of the procedure

Table 2. The fitted widths obtained from Gaussian fits to the spectra. Column 2 contains the mean value of the Gaussian standard deviation fitted to spectra collected with a gold analyser. Column 3 contains the mean of data collected with the uranium analyser. Columns 4 and 5 list the corresponding atomic kinetic energies in K. Rows with (*) symbols refers to cooled U filter data (see text). Column 6 lists the estimated condensate fraction n_0, obtained as described in the text.

Temperature (K)	σ(Au) (Å^{-1})	σ(U) (Å^{-1})	κ(Au) (K)	κ(U) (K)	n_0
1.3	0.875 ± 0.008	0.881 ± 0.011	13.9 ± 0.3	14.1 ± 0.4	15 ± 4
1.7	0.889 ± 0.009	0.899 ± 0.011	14.4 ± 0.3	14.7 ± 0.4	12 ± 4
1.9	0.903 ± 0.009	0.907 ± 0.011	14.8 ± 0.3	15.0 ± 0.4	10 ± 4
2.5	0.963 ± 0.009	0.945 ± 0.011	16.9 ± 0.3	16.2 ± 0.4	0
1.5		—		13.5 ± 0.7	11.6 ± 3.3
2.5		0.935 ± 0.019		15.9 ± 0.7	0

of fitting a single Gaussian, data were simulated using the values for double Gaussian fits tabulated in [8], table 1, and then fitted with a single Gaussian. It was found that the single-Gaussian fit produced a systematically lower kinetic energy than the sum of the two Gaussians. For example, at 1.5 K an input kinetic energy of 14.4 K was fitted as 13.3 K, whereas at 2.3 K the input value of 16.1 K was fitted as 15.4 K. Thus this simple data analysis procedure may introduce a systematic error ~ 1 K in the derived kinetic energy, but will not significantly affect the measured rate of change of kinetic energy with temperature.

In the case of the ^{4}He data at 1.5 K and 2.5 K recorded with a cooled U filter a different analysis was performed, since the better resolution made analysis of the line shape feasible. Experimental data were symmetrized with respect to $y = 0$ in order to remove the antisymmetric contribution to FSEs. The latter is, according to the most recent theoretical description [29], the only non-negligible component in ^{4}He around $q = 150$ Å^{-1}. For normal ^{4}He (2.5 K) the kinetic energy was obtained using a functional form [11] containing a fixed non-Gaussian component described by a kurtosis parameter, $\delta = 0.63$, obtained from previous PIMC [27] and experiments [30, 31]. Figure 3(a) shows the data and fit, while figure 3(b) shows the Gaussian and non-Gaussian contributions. In the case of superfluid ^{4}He at 1.5 K the kinetic energy and condensate fraction were estimated using the well known modified Sears model [32]. This assumes that the overall momentum distribution $n(p)$ has the form

$$n(p) = n_0[\delta^3(p) + f(p)] + A_1 n^*(p) \tag{6}$$

where n_0 is the condensate fraction, $n_0 f(p)$ is the condensate induced component in the $p > 0$ states, and $A_1 n^*(p)$ is the remaining distribution above the condensate and is similar to that in normal ^{4}He, with A_1 a proper constant defined by the normalization of the overall $n(p)$. The fit of the $T = 1.5$ K data was performed using a model $J(y)$ derived from the previous $n(p)$ by means of (3) and enlarged by instrumental resolution. Coefficient n_0 was considered as a free parameter. The result of this fit is shown in figure 4(a), while figure 4(b) shows the two $J(y)$ components derived from $A_1 n^*(p)$ and $n_0[\delta^3(p) + f(p)]$, respectively. The value of the kinetic energy obtained by this procedure is essentially determined by A_1 times the kinetic energy of the normal liquid ^{4}He just above the λ transition since the other two terms do not contribute significantly. These values are listed in table 2 together with the estimated condensate fraction, n_0.
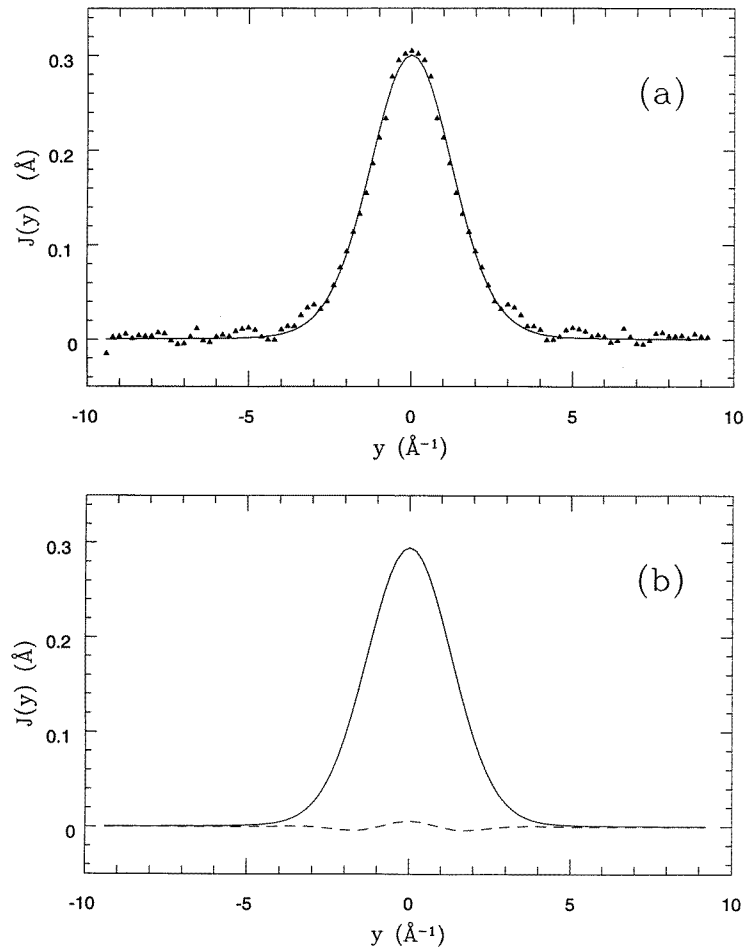
Figure 3. (a) The response function $J(y)$ for ^{4}He at 2.5 K from the cooled U filter: triangles are the experimental data; the solid line is the result of the fit. (b) The solid line is the Gaussian component of the fit; the dashed line is the non-Gaussian component, with both including a resolution contribution.

4. Conclusions and discussion

Measurements of the kinetic energy in ^{4}He have been performed by neutron Compton scattering at momentum transfers in excess of 140 Å^{-1}. The measurements show a systematic change in kinetic energy with decreasing temperature, which is consistent with the formation of a Bose condensate fraction of $\sim$10% in the liquid. The advantages of the measurements described here is that they have a high count rate and deviations from the impulse approximation are negligible. Thus the kinetic energy can be determined with a minimum of theoretical assumptions.

In figure 5 we show the values of kinetic energy obtained at different temperatures and using different resolutions and analysis procedures. These are also compared with path integral Monte Carlo calculations [33] and previous measurements [9] performed at lower momentum transfers. There is good general agreement between the eVS measurements and
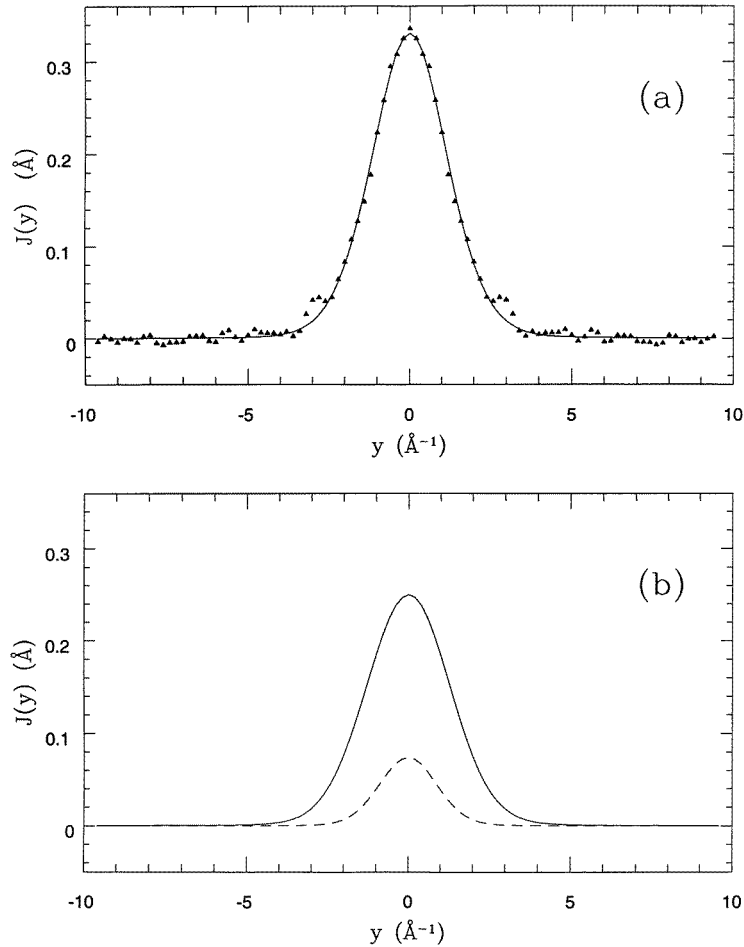
Figure 4. (a) The response function $J(y)$ for ^{4}He at 1.5 K from the cooled U filter: triangles are the experimental data; the solid line is the result of the fit. (b) The solid line is the normal fluid component of the fit; the dashed line is the Bose condensate component, with both including a resolution contribution.

previous measurements and theoretical calculations, although we note that there is some indication in figure 4 that PIMC calculations in the normal fluid give a lower kinetic energy than both our and previous experimental data.

The agreement between the gold foil and uranium foil measurements is very good, despite the large differences in the width and shapes of the resolution functions. This suggests that the energy analysis using a gold foil does give accurate values of kinetic energy, despite the fact that the measurements are very resolution limited. Moreover the kinetic energies obtained from the cooled U filter ^{4}He data are consistent with values obtained using the simpler analysis procedure, which suggests that the use of a single Gaussian in the fitting procedure does not introduce any serious error in the values of kinetic energy derived from the fits.

In contrast to earlier neutron work at eV energy transfers [13], the measurements suggest a condensate fraction close to 10%, in agreement with the best theoretical estimates and with
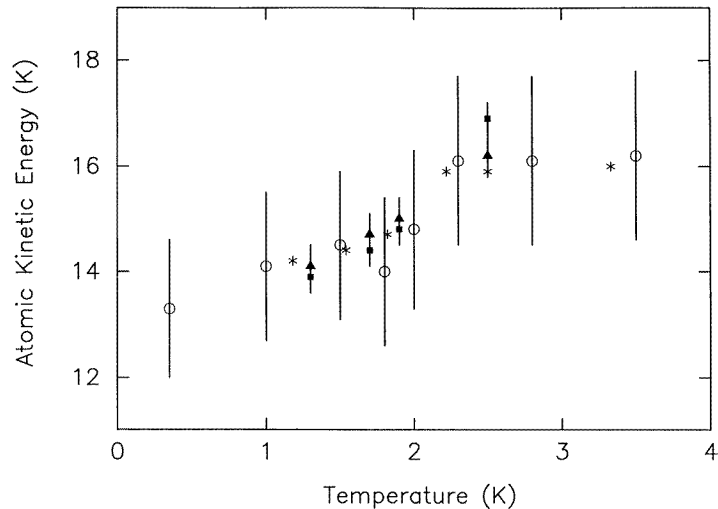
Figure 5. Shows a comparison of kinetic energy values listed in table 2 with previous work. The solid triangles are eVS data taken with the uranium analyser foil. The solid squares are eVS data taken using the gold foil. The circles are values measured previously by Sokol and coworkers [9] and the symbols * give values calculated by path integral Monte Carlo techniques [33]. The error bars on the points are purely statisical and do not include possible systematic errors.

previous work at lower momentum transfers. The more sophisticated analysis performed on the data taken with a cooled uranium foil is consistent with the hypothesis that the atomic momentum distribution in the normal fluid contains a small non-Gaussian component, whereas in the superfluid this distribution appears strongly non-Gaussian.

The improvement in resolution and the development of sensitivity to line shape, which occurs in the data taken with a cooled uranium foil, shows that there is much to be gained by improving the resolution in eV measurements. Gains in resolution by a factor of two are obtainable by cooling the analyser foil and increasing the instrument flight paths. Future developments in eV neutron scattering will provide comparable resolution to chopper spectrometers, but with much higher count rates and momentum transfers a factor of six greater. It will then be possible to perform more detailed line shape analysis on ^{4}He data and perhaps obtain the long-sought direct evidence for the presence of a Bose condensate fraction in superfluid ^{4}He.

Acknowledgments

References

[1] Hohenberg P C and Platzmann P M 1966 *Phys. Rev.* **152** 198
[2] West G B 1989 *Momentum Distributions* ed R N Silver and P E Sokol (New York: Plenum) p 95

[3] Sears V F 1984 *Phys. Rev.* B **30** 44
[4] Mayers J 1990 *Phys. Rev.* B **41** 41
[5] Mayers J, Andreani C and Baciocco G 1989 *Phys. Rev.* B **39** 2022
[6] Watson G 1996 *J. Phys.: Condens. Matter* **8** 5955
[7] Mook H A 1983 *Phys. Rev. Lett.* **51** 1454 and references therein
[8] Sosnick T R, Snow W M, Sokol P E and Silver R N 1989 *Europhys. Lett.* **9** 707
[9] Sosnick T R, Snow W M, Sokol P E and Silver R N 1990 *Phys. Rev.* B **41** 11 185
[10] Silver R N 1987 *Phys. Rev.* B **37** 3794 and references therein
[11] Glyde H R 1994 *Phys. Rev.* B **50** 6726
[12] Azuah R T, Stirling W G, Glyde H R, Sokol P E and Bennington S M 1995 *Phys. Rev.* B **51** 605
[13] Ikeda S and Watanabe N 1987 *Phys. Lett.* **121A** 34
[14] Mayers J 1996 *J. Phys.: Condens. Matter* **8** 9423. Contains a review of quantum fluids measurements on
     eVS.
[15] Andreani C, Filabozzi A, Nardone M, Ricci F P and Mayers J 1994 *Phys. Rev.* B **50** 12 744
[16] Bafile U, Zoppi M, Barocchi F, Magli R and Mayers J 1996 *Phys. Rev.* B **54** 11 969
[17] Bafile U, Zoppi M, Barocchi F, Magli R and Mayers J 1995 *Phys. Rev. Lett.* **75** 1957
[18] Azuah R T, Stirling W.G, Mayers J, Bailey I F and Sokol P E 1995 *Phys. Rev.* B **51** 6780
[19] Timms D N, Evans A C, Boninsegni M, Ceperly D M, Mayers J and Simmons R O 1996 *J. Phys.: Condens.
     Matter* **8** 6665
[20] Mayers J 1993 *Phys. Rev. Lett.* **71** 1553
[21] Mayers J, Burke T M and Newport R N 1994 *J. Phys.: Condens. Matter* **6** 641. This paper contains a detailed
     account of eVS and the data analysis procedures used.
[22] Fillaux F, Baron M H, Mayers J and Tomkinson J 1995 *Chem. Phys. Lett.* **240** 114
[23] Evans A C, Mayers J and Timms D N 1996 *Phys. Rev.* B **53** 3023
[24] Fielding A L, Timms D N, Evans A C and Mayers J 1996 *J. Phys.: Condens. Matter* **8** 7205
[25] Mitchell P C H, Green D A, Payen E and Evans A C 1995 *J. Chem. Soc. Faraday Trans.* **91** 4467
[26] Bermejo F J, Mompean F J, Mayers J and Evans A C 1994 *Phys. Lett.* **189A** 333
[27] Andreani C, Baciocco G, Holt R S and Mayers J 1989 *Nucl. Instrum. Methods* A **276** 297
[28] Azuah R T, Stirling W G, Glyde H R, Sokol P E and Bennington S M 1995 *Phys. Rev.* B **51** 605
[29] Rinat A S, Taragin M F, Mazzanti F and Pools A 1997 *Phys. Rev.* B submitted
[30] Andersen K H, Stirling W G, Glyde H R, Azuah R T, Taylor A D, Bennington S M and Bowden Z A 1994
     Physica B **197** 198
[31] Andersen K H, Stirling W G, Taylor A D, Bennington S M, Bowden Z A, Bailey I and Glyde H R 1992
     Physica B **180/181** 865
[32] Sears V F, Svensson E C, Martel P and Woods A D B 1982 *Phys. Rev. Lett.* **49** 279
[33] Ceperley D M and Pollock E L 1987 *Can. J. Phys.* **65** 1416