Measurement of atomic momenta in quantum fluids by high-energy neutron scattering

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Abstract. This paper gives a brief outline of the theory and practice of kinetic energy measurements made on the electronvolt spectrometer at the ISIS spallation neutron source. The usefulness of the technique is illustrated by measurements made on ^{4}He as a function of temperature and density, on ^{3}He–^{4}He mixtures as a function of concentration and on liquid neon. Future developments and applications are discussed.

The measurement of atomic momentum distributions in condensed-matter systems by high-energy neutron scattering is analogous to the measurement of electron momentum distributions by Compton scattering and the measurement of nucleon momenta by deep inelastic scattering [1]. The technique is known as neutron Compton scattering (NCS) or deep inelastic neutron scattering (DINS) by analogy with these older techniques. The impulse approximation (IA), which is exact only in the limit of infinite momentum transfer [2, 3], is used in the interpretation of NCS measurements, and large incident energies are required to keep deviations from the IA small.

NCS measurements on most systems have only become feasible with the development of the new generation of pulsed neutron sources, such as ISIS at the Rutherford Appleton Laboratory, UK. For accurate NCS measurements, energy transfers about 100 times greater than the mean atomic kinetic energy are required and only pulsed neutron sources can supply a sufficiently high intensity of neutrons at these energies. Many early NCS measurements were performed on helium at relatively low energy and momentum transfers ($\omega < 300$ meV and $q < 15$ Å^{-1}) at reactor sources. These studies were motivated primarily by the possibility of directly observing the Bose condensate fraction in superfluid ^{4}He. However, despite the low kinetic energy of ^{4}He atoms (about 1.5 meV), there are still significant deviations from the IA due to 'final-state' broadening of the expected δ-function component in the momentum distribution [2] and higher neutron energies are therefore desirable. More recently NCS measurements with 15 Å$^{-1} < q < 40$ Å^{-1} and incident energies up to 2 eV have been made on condensed phases of helium [4] and neon [5].

The electronvolt spectrometer at ISIS [6, 7] has been dedicated to the development of the NCS technique during the last 5 years. The unique attributes of the electronvolt spectrometer are very high momentum transfers (1 Å$^{-1} < q < 150$ Å^{-1}) and energy transfers (1 eV $< \omega < 50$ eV) two orders of magnitude greater than those previously accessed by neutron scattering. At such high q and ω the IA is a good approximation both in quantum fluids and in more strongly bound systems. There are many applications of NCS in areas other than the study of quantum fluids, e.g. hydrogen bonds [8, 9], polymers [10], glasses [11, 12], amorphous materials [13], metals [14] and metal hydrides [15].

The IA [2, 3] states that, if the momentum lost by the incident neutron is sufficiently large, scattering occurs from a single atom with conservation of kinetic energy and momentum. Consider a scattering event in which a neutron loses momentum q and energy ω scattering from an atom of mass M. If the momentum of the atom is p before the collision, then from conservation of momentum it is $p + q$ after the collision and to conserve kinetic energy the equation

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

must be satisfied. Rearrangement of this equation gives

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

where $\hat{q} = q/|q|$ is the unit vector along the direction of q and y is the component of atomic momentum along the direction of q. In fluids the direction is immaterial and we measure $J(y)$, the distribution of momentum components, along an arbitrary direction in space. The atomic kinetic energy is related to $J(y)$ via

$$\kappa = \frac{3}{2M} \int_{-\infty}^{\infty} y^2 J(y) \, \mathrm{d}y. \tag{3}$$

The electronvolt spectrometer uses a filter difference method [16] to analyse the energy of the scattered neutrons. The filter is a thin foil of either gold or uranium, which absorbs neutrons strongly over narrow bands of energy, centred at energy E_1. Two time-of-flight spectra are taken, one with the foil between the sample and detectors and the second with the foil removed. The difference between these spectra is due to neutrons absorbed in the foil and is effectively the time-of-flight spectrum for neutrons scattered with energy E_1. The gold foil gives a Lorentzian energy resolution function, centred at $E_1 = 4.908$ eV with a half-width at half-maximum (HWHM) of 0.138 eV, whereas the uranium foil gives a Gaussian energy resolution function centred at $E_1 = 6.671$ eV and with a standard deviation of 0.063 eV. If E_1 is known, the momentum transfer q and energy transfer ω can be determined from a measurement of the neutron time of flight t in the usual way [17], and hence from equation (2) the atomic momentum component y can be found.

The data analysis procedures for quantum fluids using the electronvolt spectrometer consists in fitting a Gaussian peak shape convolved with the instrument resolution function. The standard deviation σ of the Gaussian fit is related to the mean atomic kinetic energy via $\kappa = 3\sigma^2/2M$. The instrument resolution is determined by a standard calibration procedure, the accuracy of which has now been extensively checked both by its internal consistency and by the good agreement obtained with calculations on systems which are well understood or with previous measurements. An example of the accuracy of the procedure is given by results obtained on liquid helium. Two simultaneous measurements were made using both uranium and gold foil analysers. Although in the latter case the resolution function is much broader than the intrinsic width of $J(y)$, the measurements are in good agreement with each other, as can be seen from table 1.

Despite the currently limited resolution, electronvolt spectroscopy still has very significant advantages over measurements performed at lower q and ω. The count rates are higher by one to two orders of magnitude, deviations from the IA are small and the signal from the sample is well separated from that produced by the sample cell. There is considerable interest in determining how the kinetic energy of atoms in quantum fluids changes as a function of thermodynamic parameters such as temperature, pressure and sample composition.

Table 1. Measured kinetic energies in liquid ^{4}He as a function of temperature made using gold and uranium foil analysers [30]. In the former case the energy resolution function is Lorentzian in shape with a HWHM of 1.9 Å^{-1} while in the latter case it is Gaussian with a standard deviation of 0.76 Å^{-1}. The extracted standard deviation of the Gaussian fit to the momentum distribution is about 0.9 Å^{-1}. Despite the widely different resolution functions, good agreement between the two measurements is obtained, indicating that the resolution calibration is accurate.

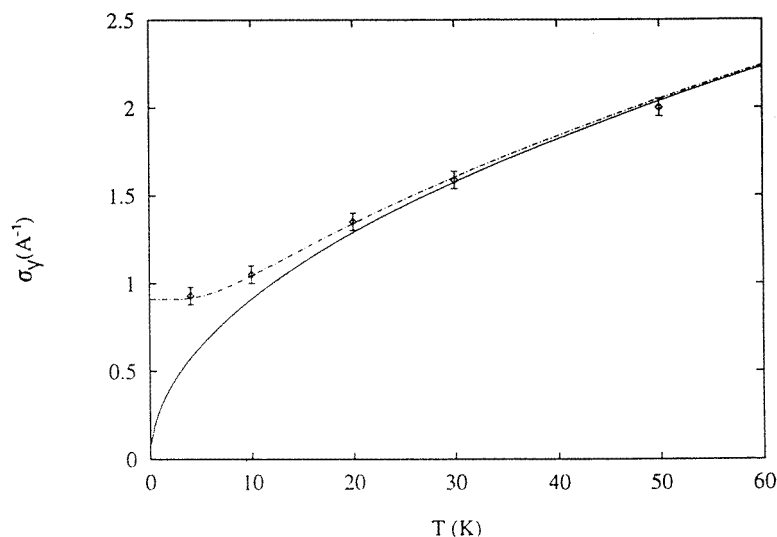|	Kinetic energy (K)	
Temperature (K)	Au foil analyser	U foil analyser
1.3	13.9 ± 0.3	14.1 ± 0.4
1.7	14.4 ± 0.3	14.7 ± 0.4
1.9	14.8 ± 0.3	15.0 ± 0.4
2.5	16.9 ± 0.3	16.2 ± 0.4

Figure 1. Standard deviation σ of Gaussian fit to data on ^{4}He at a constant density of 0.15 g cm^{-3}, as a function of temperature: ——, classical limit; — · —, fit using the Einstein model.

Figure 1 shows a measurement of the kinetic energy of ^{4}He as a function of temperature at constant density [18]. The residual zero-point motion of the atoms at low temperatures is clearly seen. Figure 2 shows a similar measurement [19] performed as a function of density at $T = 4.35$ K. The slope of the curve changes significantly at the liquid–solid transition and there appears to be a discontinuity in the kinetic energy. Figure 3 shows the kinetic energy of ^{3}He atoms in a ^{3}He–^{4}He mixture, measured as a function of ^{3}He concentration [20]. The experiment is in good agreement with previous measurements using chopper spectrometers but disagrees with theory. The latter predicts that the kinetic energy decreases as the concentration of ^{3}He increases, but this is not observed. Recently measurements on liquid neon [21] have yielded precise information about the temperature dependence of kinetic energies, allowing a sensitive test of modern theoretical calculations of the properties of the liquid.
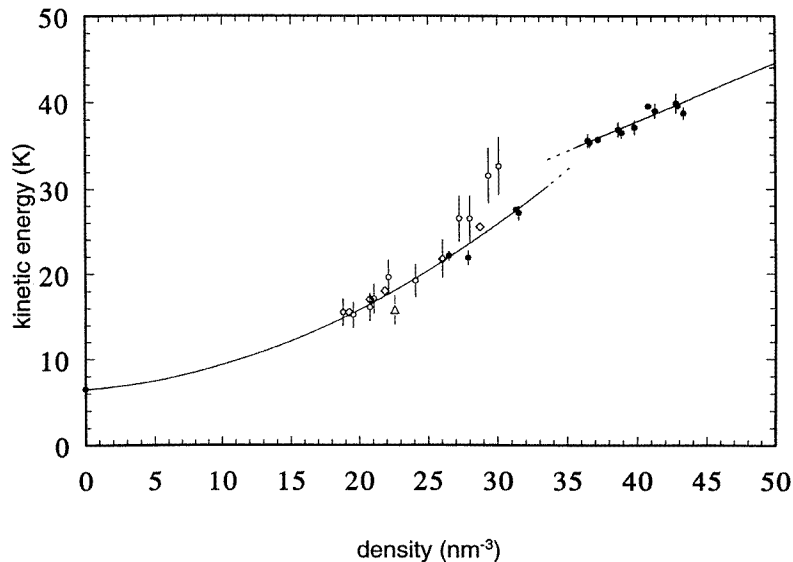
Figure 2. The kinetic energy of liquid and solid ^{4}He as a function of density at $T = 4.35$ K: ● , measured on the electronvolt spectrometer; ○ , data obtained from [24]; △, datum obtained from [18]; ◊, the path integral Monte Carlo calculations given in [24]; the point at zero density is the classical value ($3 kT/2$); ——, polynomial fits to the data.
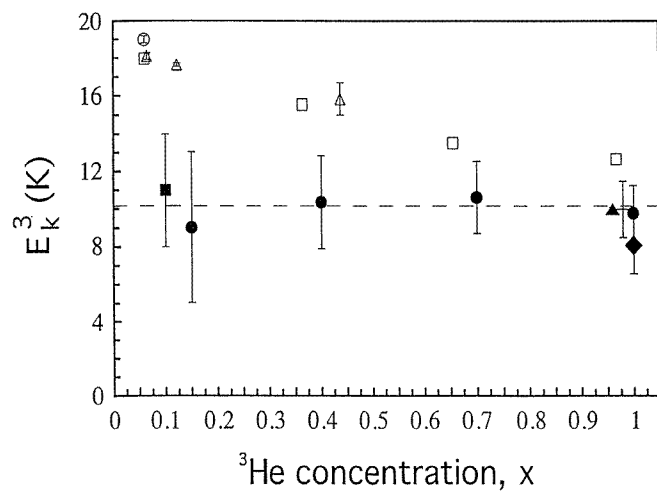

Figure 3. Concentration dependence of the ^{3}He kinetic energy in a ^{3}He–^{4}He mixture: ● , electronvolt spectrometer results; ■, experimental result from [25]; ♦, experimental result from [26]; ○ , calculation from [27]; △, calculation from [28]; □, calculation from [29]. There is clear disagreement between experiment and theory.

Another interesting application of electronvolt spectroscopy was to the measurement of the proton wavefunction in liquid hydrogen [22]. Although the measured $J(y)$ is dominated by the internal binding of the H_2 molecule, a difference [23] in the profiles could be observed in the liquid and the solid, owing to the different molecular centre-of-mass motions in the

two systems. This suggests the possibility of observing both the internal and the centre-of-mass motion in molecular liquids.

The technique of using electronvolt neutrons to measure atomic momentum distributions is still in its infancy. The resolution width could be improved by a factor of 2 by cooling the analyser foils and increasing the instrument flight paths, and future measurements will certainly involve a more detailed analysis of the peak shape of $J(y)$ in quantum fluids. This can be expected to yield much more detailed information on the atomic dynamics. Large increases in count rate can also be obtained by increasing the modest detector area currently available. However, as the above examples demonstrate, the technique provides unique information about the dynamics of quantum fluids under a variety of conditions and is already showing much promise.

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