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Momentum and energy dependent resolution function of the ARCS neutron chopper spectrometer at high momentum transfer: Comparing simulation and experiment

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ABSTRACT

Inelastic neutron scattering at high momentum transfers (i.e. $Q \geq 20$ Å), commonly known as deep inelastic neutron scattering (DINS), provides direct observation of the momentum distribution of light atoms, making it a powerful probe for studying single-particle motions in liquids and solids. The quantitative analysis of DINS data requires an accurate knowledge of the instrument resolution function $R_i(Q, E)$ at each momentum Q and energy transfer E, where the label i indicates whether the resolution was experimentally observed $i = obs$ or simulated $i{=}sim$. Here, we describe two independent methods for determining the total resolution function $R_i(Q, E)$ of the ARCS neutron instrument at the Spallation Neutron Source, Oak Ridge National Laboratory. The first method uses experimental data from an archetypical system (liquid ^{4}He) studied with DINS, which are then numerically deconvoluted using its previously determined intrinsic scattering function to yield $R_{obs}(Q, E)$. The second approach uses accurate Monte Carlo simulations of the ARCS spectrometer, which account for all instrument contributions, coupled to a representative scattering kernel to reproduce the experimentally observed response $S(Q, E)$. Using a delta function as scattering kernel, the simulation yields a resolution function $R_{sim}(Q, E)$ with comparable lineshape and features as $R_{obs}(Q, E)$, but somewhat narrower due to the ideal nature of the model. Using each of these two $R_i(Q, E)$ separately, we extract characteristic parameters of liquid ^{4}He such as the intrinsic linewidth α_2 (which sets the atomic kinetic energy $\langle K \rangle \sim \alpha_2$) in the normal liquid and the Bose–Einstein condensate parameter n_0 in the superfluid phase. The extracted α_2 values agree well with previous measurements at saturated vapor pressure (SVP) as well as at elevated pressure (24 bars) within experimental precision, independent of which $R_i(Q, y)$ is used to analyze the data. The actual observed n_0 values at each Q vary little with the model $R_i(Q, E)$, and the effective Q-averaged n_0 values are consistent with each other, and with previously reported values.

1. Introduction

Due to the unique properties of the neutrons, their use as an experimental probe for studying atomic (or molecular) vibrations and interactions is a rather well-established technique which has contributed to many advances in various scientific areas such as condensed matter physics and chemical sciences [1,2]. Notable recent examples have revealed the existence of a magnetic resonance peak in iron-based superconductors [3], clarified the connection between Bose–Einstein condensation and superfluidity in the Bosonic liquid ^{4}He [4], unravelled the existence of a roton-like signature in non-Bosonic liquid ^{3}He [5], found a non-Gaussian proton momentum distribution in confined water [6,7], etc. The momentum Q and energy E landscape that can be probed by neutrons is rather broad, and generally not accessible by a single instrument and multiple reconfigurations of the same instrument [8] are often necessary. This is because each neutron instrument is tailored and optimized (by design and technical limitations) to only probe a small cross-section of the wider Q–E space, making it often necessary to use multiple neutron instruments with overlapping Q–E windows and/or other complementary techniques before a particular phenomenon can be fully understood. One notoriously known limiting factor for reconciling and interpreting neutron scattering data from different instruments is the fact that

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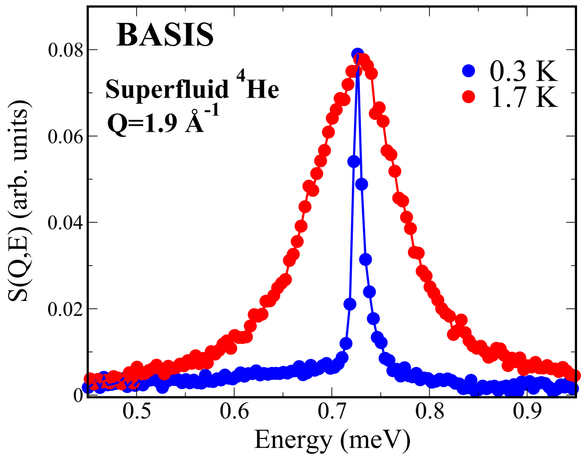

Fig. 1. Temperature dependence of the roton mode in superfluid ^{4}He, as observed on the BASIS neutron spectrometer at the Spallation Neutron Source. The resolution limited asymmetric sharp peak (blue circles) at low temperature is used to represent the intrinsic energy resolution function (HWHM ~5.8 μeV) for studying the temperature dependence of the roton lifetime at T=1.7 K (red circles). (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper.)

the energy resolution function of a given instrument depends not only on the instrument parameters but also on the imparted momentum Q and energy E to the sample. Therefore, while qualitative interpretation of the raw INS data is generally possible (peak positions, dispersive nature of the excitations, etc.), any rigorous quantitative analysis of INS data (excitation lifetimes, BEC fraction, etc.) requires an accurate knowledge of the resolution function $R_i(Q, E)$ [8–11].

To set the background, we note that at small momentum transfers Q (i.e. $Q \le 2$ Å^{-1}), inelastic neutron scattering (INS) is quite effective in examining molecular re-orientation, diffusion processes and low energy excitations. In those cases, the resolution function is usually obtained by directly measuring the exact same sample at the lowest possible temperatures where dynamical processes become frozen out on the instrument measurement time window, leaving out only the instrument contributions at or close to the elastic region. This is particularly true for backscattering instruments where the energy resolution width remains largely constant over the accessible Q range and over a fairly broad dynamics range close to the elastic peak [12]. Fig. 1 compares the observed elementary excitation (or 'roton') in superfluid ^{4}He at temperature of 1.7 K to its resolution limited response at 300 mK on the BASIS neutron spectrometer [13] at the Oak Ridge National Laboratory's Spallation Neutron Source (SNS). The low temperature measurement faithfully reproduces the asymmetric resolution due to the liquid H moderator source. On most indirect geometry neutron instruments such as BASIS, or direct geometry instruments in low Q mode such as the DCS [14] at the NIST center for neutron research, it may at times be sufficient to simply measure a purely incoherent standard such as vanadium in lieu of the low temperature measurements to determine $R_i(Q, E)$. Unfortunately, no such straightforward practical approach exists when dealing with INS data from direct geometry instruments at high Q's. At intermediate Q's (i.e. $1 \le Q \le 10$ Å^{-1}), which is well-suited for studying collective excitation modes such as phonons and magnons, and characteristic excitations such as rotons and molecular crystal fields, there exist few benchmarked software tools that account for resolution contributions in analyzing INS data collected on time-of-flight chopper spectrometers. The TOBYFIT software package [15] which uses a semi-empirical method to approximate the resolution function has been the primary workhorse for analyzing time-of-flight INS data in this regime. On the other end of the Q spectrum, i.e. the high momentum transfer regime ($Q \le 20$ Å^{-1}), which is the subject of this study, the inelastic scattering process of the individual atoms resembles closely the scattering of freely moving particles and the neutron response is characterized by recoil scattering. In these two cases, $R_i(Q, E)$ can either be inferred from measuring calibrated samples for which the scattering function is well known so that it can be numerically deconvoluted from the measurements, or from using ray tracing Monte Carlo methods with all instrument characteristics as input. In this article, we present the resolution function of the ARCS neutron spectrometer at the Spallation Neutron Source [16] at the Oak Ridge National Laboratory obtained using each of these two procedures. The main goal is to properly account for the instrument resolution contributions when analyzing INS data obtained at high Q's (i.e. DINS). Using these differently obtained $R_i(Q, E)$, we investigate the changes in the resulting average kinetic energy $\langle K \rangle$ of the atoms in normal liquid ^{4}He, and the fraction of atoms that Bose condense (BEC) in the superfluid phase. While $\langle K \rangle$ is directly proportional to the linewidth α_2 of the DINS signal, the macroscopic number of atoms in the BEC state can be inferred from the relative change in intensity in the DINS response between the normal and superfluid phases. Our analysis shows consistent $\langle K \rangle$ results with previous measurements at saturated vapor pressure (SVP) [17] and at elevated pressure near the liquid–solid transition line [18]. The condensate fraction n_0 shows however sensitivity to otherwise marginally different $R_i(Q, E)$.

2. Neutron measurements

2.1. ARCS: the wide-angle neutron chopper spectrometer

The wide angular range chopper spectrometer ARCS [16,19] is one of four direct geometry neutron instruments located at the Spallation Neutron Source (SNS), Oak Ridge National Laboratory. This means that it uses a monochromatic beam of neutrons whose energy E_i can be set by the experimenter by varying the rotation speed and phase of a spinning Fermi chopper [20] located before the sample position to probe large area of momentum–energy (Q–E) space of a subject material. A model view of the ARCS instrument layout is illustrated in Fig. 2 for more details. The final energy E_f of the neutrons after scattering from the sample is determined by time-of-flight techniques (TOF), allowing the energy transfer E to the sample to be calculated, $E = E_i - E_f$. Using kinematic constraints for a given incident energy E_i, the momentum transfer Q can be conveniently expressed as a function of the energy transfer E, and the scattering angle ϕ, i.e. the angle between the incident and scattered beam (often called 2θ in diffraction methods), yielding,

$$Q^2 = \frac{1}{\gamma}\left(2E_i - E - 2\sqrt{E_i(E_i - E)}\cos\phi\right) \tag{1}$$

where $\gamma = \frac{\hbar^2}{2m_n} = 2.017$ meV Å^2. On ARCS, this parametric relation allows large region of Q–E to be probed, thanks to the large ϕ coverage by the detector arrays, $-28° \le \phi \le 135°$.

Fig. 3 illustrates the large $Q - E$ range accessible on ARCS with an incident energy E_i of 686 meV, as used in our experiment. The top panel shows the original Q–E contour plot from bulk liquid ^{4}He at saturated vapor pressure (SVP). As can be appreciated, this raw data is dominated by the ^{4}He signal (bright parabolic strip), but does include elastic contributions (strip around E=0), recoil scattering from other heavier elements (mostly Al, marked by the broad low energy parabolic band), and other intrinsic background contributions (diffuse signal). The bottom panel shows the