Quantum and classical behavior of single-particle dynamics in dense liquid ^{4}He

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Inelastic-neutron-scattering measurements at high exchanged momentum have been performed in fluid ^{4}He along a supercritical isochore in the 4.2–50 K temperature range. The data have been analyzed in the plane-wave impulse approximation to obtain the root-mean-square values of the particle momentum. Deviations from classical behavior have been interpreted as arising from the quantum nature of the particle oscillations in the framework of a cell model for the fluid.

I. INTRODUCTION

Dense liquid helium is of great interest to fluid state physicists since it represents the prototype system of a quantum fluid. In this system much experimental effort has been devoted to the study both structural[1] and dynamical[2,3] properties for thermodynamic states along the liquid side of the liquid-vapor saturation line and across the λ transition. On the contrary less attention has been devoted to the understanding of the changes that occur in the dynamical properties as the system goes from the classical to the quantum regime.

In this context an interesting dynamical quantity is the single-particle momentum distribution, $n(p)$, which determines, for instance, the value of the average kinetic energy $\langle E_k \rangle$. In the last decade experimental measurements of $n(p)$ have become available from inelastic-neutron-scattering experiments performed at high-momentum transfer. Using this technique, measurements of $n(p)$ have already been performed in low-temperature liquid ^{4}He as a function of density.[4] At the investigated temperature (4.2 K) the quantum nature of the fluid is clearly evident being the variance of $n(p)$ much larger than the classical value Mk_BT.

In this work inelastic-neutron-scattering measurements from fluid ^{4}He are reported along a supercritical isochore at a density close to that of the lambda transition up to a temperature of 50 K where the system is expected to behave classically.

II. EXPERIMENT

Measurements of the dynamical structure factor $S(q,\omega)$ have been performed using eVS spectrometer at ISIS, where the inelastic-neutron-scattering pattern of the sample is determined using the filter difference technique.[5] A gold foil is the filter employed which ensures an adequate intensity and resolution for samples of relatively low masses, and presents just one nuclear resonance. Ten scintillator detectors were placed in the angular range $110° < \theta < 150°$ corresponding to a momentum transfer ranging from 97 to 124 A^{-1}. A more detailed description of the experimental feature of eVS spectrometer can be found in Ref. 5. The ^{4}He sample was contained in a cylindrical aluminum can (inner diameter $\cong$11 mm, outer diameter $\cong$13 mm). The aluminum can ensures that the signal from the container results well separated in energy from that coming from the sample as well as providing a relatively small scattering contribution. Five measurements on the 0.15 ± 0.01 g/cm^3 isochore were performed in the 4.2–50.0 K temperature range thus implying a maximum working pressure of 233 bar.

A typical differenced time-of-flight spectrum from one detector is plotted in Fig. 1 for $T=50$ K. The recoil peak intensity of the sample at each angle occurs at smaller time of flight then that from the can. The two peaks remain well separated in time of flight in the whole angular range. The large values of momentum transfer, q, allow us to describe the scattering in terms of the plane-wave impulse approximation (PWIA).[6] In this framework the ω and q variables are coupled through the appropriate scaling variable $y=(M/\hbar q)(\omega-\hbar q^2/2M)$

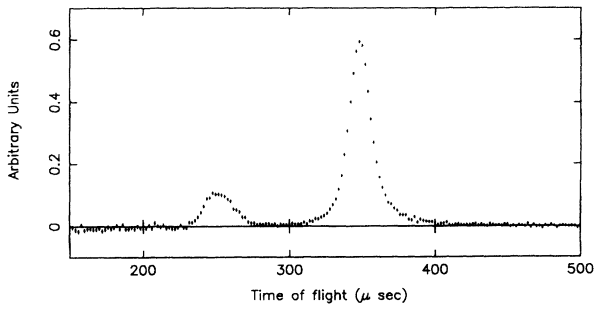

FIG. 1. Differenced spectrum from one detector plotted as a function of time of flight for $T=50$ K.

and the scaling function $F(y)=(\hbar q/M)S(q,\omega)$ can be derived for each angle.

For each temperature, these functions have been fitted, at each scattering angle, accounting for the finite experimental resolution,[5] using a Gaussian line shape. The variance σ_y of these Gaussians, which is the only fit parameter, is found to be independent on the scattering angle. Therefore, the average value of σ_y for each temperature is reported. These values, which represents the root-mean-square value of a Cartesian component of the particle momentum in $\hbar$ units, are listed in Table I together with the corresponding kinetic energies $\langle E_K \rangle = 3\hbar^2\sigma_y^2/2M$. In Fig. 2, the scaling functions $F(y)$ obtained averaging the experimental results over the scattering angles for the five investigated temperatures are reported together with the best-fit line shapes and with the instrumental resolution adopted. Temperature broadening effects are clearly visible in this figure.

TABLE I. Variances σ_y of the scaling function $F(y)$ and corresponding average kinetic energies at the temperatures of the experiment.

T (K)	σ_y (A^{-1})	$\langle E_K \rangle$ (K)
4.2	0.93±0.05	15.8±1.7
10.0	1.05±0.04	20.1±1.5
20.0	1.35±0.05	33.2±2.4
40.0	1.59±0.05	45.9±3.0
50.0	2.00±0.05	72.8±3.6

III. DISCUSSION AND CONCLUSIONS

The temperature behavior of σ_y is plotted in Fig. 3 as a function of temperature. As expected in the high-temperature region the results agree with the classical prediction $\sigma_y=\sqrt{Mk_BT_0}/\hbar^2$. At low-temperature systematic deviations from the classical behavior due to the quantum nature of the system occur yielding a finite zero-temperature limit. Unfortunately, Wigner-type expansions of the kinetic energy are hardly convergent for ^{4}He in the investigated temperature range.[7] We have, however, been able to reproduce the whole temperature dependence of σ_y using a three-dimensional isotropic Einstein oscillator model with characteristic frequency ω_0 which yields

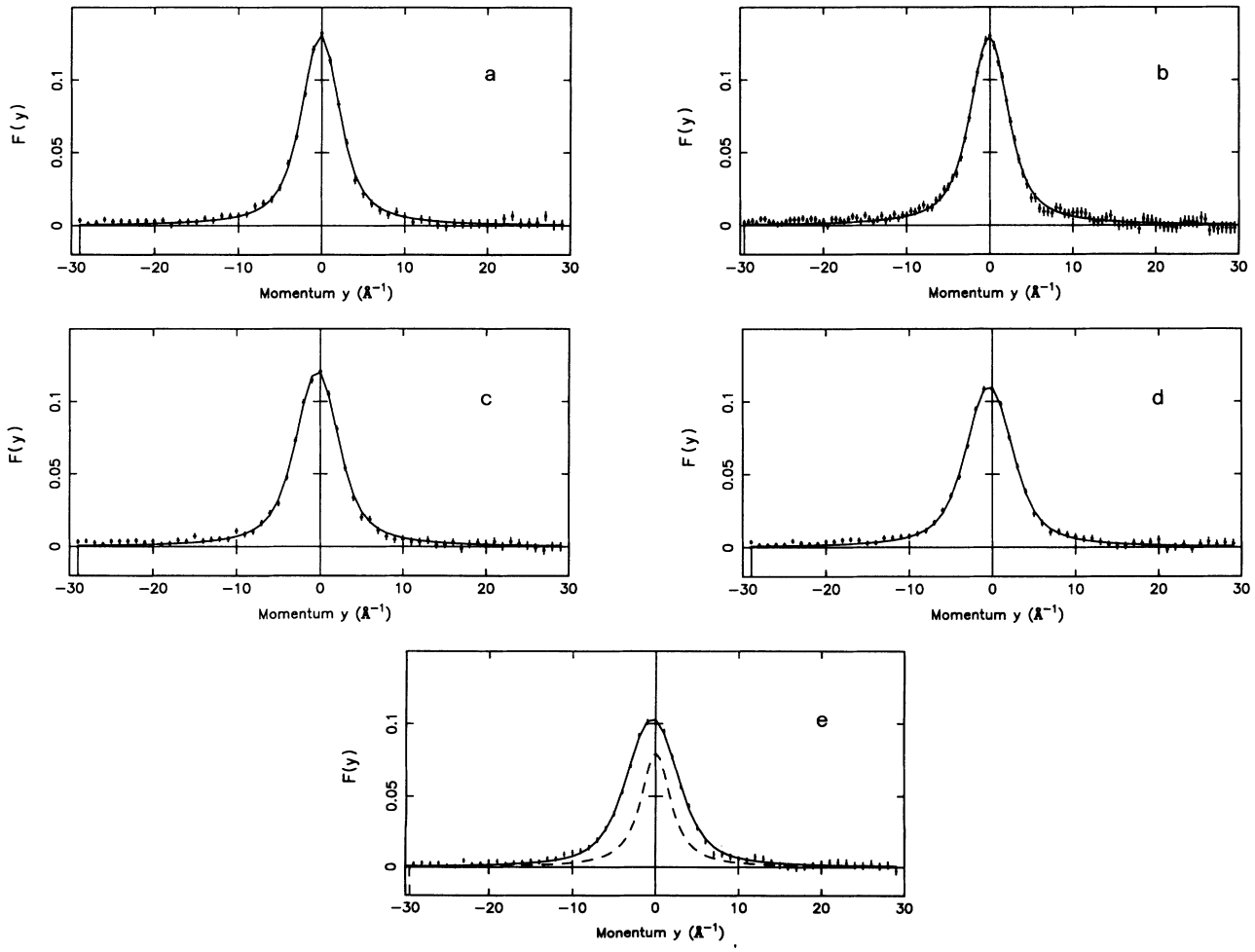

FIG. 2. The scaling functions $F(y)$ averaged over the scattering angles (dots), for the five temperatures, as a function of the scaling variable y: (a) $T=4.2$ K; (b) $T=10$ K; (c) $T=20$ K; (d) $T=30$ K; (e) $T=40$ K; (f) $T=50$ K. The solid line represents the best-fit line shape. The resolution function is also reported as a dashed line for the higher-temperature spectrum.

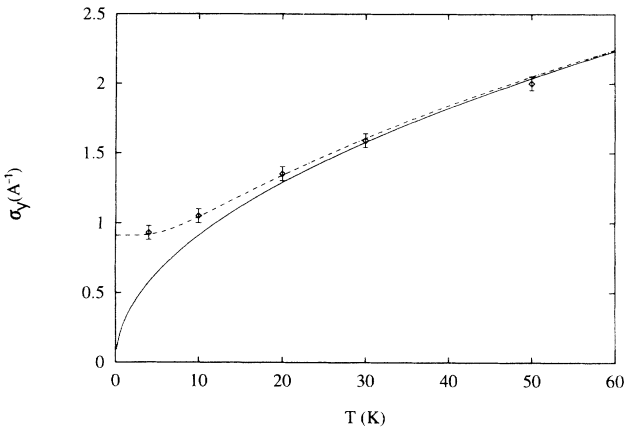

FIG. 3. The σ_y values derived from the experimental spectra are plotted as a function of temperature (dots). The solid line represents the classical limit whereas the dash-dotted line is the fitted Einstein model.

$$\sigma_y^2 = \frac{Mk_B T_0}{\hbar^2}\left[\frac{1}{e^{T_0/T}-1}+\frac{1}{2}\right], \tag{1}$$

where $T_0=\hbar\omega_0/k_B$. A good quality fit can be obtained (see Fig. 3) with $T_0=20.0\pm1.5$ K. It might be worthwhile noticing that a Debye continuum model having the same zero-point energy (i.e., a Debye temperature equal to $4/3T_0$) can parametrize equally well our data. These values correspond to a kinetic energy for the zero-point motion $\langle E_k\rangle_0=9/16\theta_D=3/4T_0=15.2\pm1.2$ K. This value is in excellent agreement with the results of path-integral Monte Carlo calculations[8] and agrees also rather well with the results of Ref. 3 that yield, at our density and at 4.2 K, a kinetic energy of about 18 ± 2 K.

The agreement found between the temperature behavior of the kinetic energy and the predictions of purely vibrational models suggest that a cell model[9] can be used in describing the average dynamical properties such as the specific heat or $\langle E_K\rangle$ in dense fluid ^{4}He. As a matter of fact if one assumes that each atom is situated in a spherical shell with a radius a over which a number of z first neighbors are uniformly distributed and that the interatomic interactions are described by a Lennard-Jones potential, with parameters ε and σ, one can easily calculate the vibrational frequency for an atom inside the cell which turns out to be

$$\omega_0=\left\{\frac{8z\varepsilon}{M\sigma^2}\left[22\left[\frac{\sigma}{a}\right]^{14}-5\left[\frac{\sigma}{a}\right]^{8}\right]\right\}^{1/2}. \tag{2}$$

Using $z=9$ and $a=3.2$ A, as obtained from the radial distribution functions of fluid ^{4}He measured at low temperature at roughly the same density,[10] $\varepsilon=14.11\times10^{-16}$ erg and $\sigma=2.56$ A^{-1},[11] one obtains $T_0=14$ K in reasonable agreement with the value obtained from the Einstein oscillator model fit to our data.

Furthermore since $a\sim\rho^{-3}$, one can see that, at densities where Eq. (2) can be applied (namely large enough to yield positive ω_0 values but still ensuring an attractive potential inside the cell) the zero-point kinetic energy behaves essentially as $\sim\rho^{2.33}$ which compares rather well with the ρ^2 behavior found experimentally.[3]

In conclusion our inelastic-neutron-scattering measurements performed at high-momentum transfer on fluid ^{4}He along a nearly isochoric path at 0.15 g/cm^3 show that, as far as single-particle average kinetic energy is concerned, quantum corrections become appreciable for $T\leq20$ K.

Its temperature behavior in the investigated range can be described in terms of purely vibrational motion of the atoms consistent with a cell model for the fluid. Such a model is also found to reproduce qualitatively the density dependence of the zero-point kinetic energy experimentally found in Ref. 3.

It will be interesting to extend these temperature-dependence measurements of σ_y to higher densities since, as indicated by the density behavior of the zero-point energy, resolution effects would be less important and more stringent tests of the theoretical models could be performed.

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