

ELEMENTARY EXCITATIONS OF LIQUID ^4He IN CONFINEMENTF. Albergamo¹, H.R. Glyde², J. Bossy³¹Laboratoire Léon Brillouin (CEA-CNRS), CEA-Saclay, 91191 Gif-sur-Yvette Cedex (France)²Department of Physics and Astronomy, University of Delaware, Newark, Delaware, 19711 (U.S.A.)³Centre de Recherche sur les Très Basses Températures, CNRS, BP 166, 38042 Grenoble (France)**1. Introduction**

Superfluidity Bose-Einstein Condensation (BEC) and extremely well-defined "phonon-roton" (p-r) excitations (full width at half maximum as narrow as 0.1 (μeV at $T = 0.8\text{ K}$) in low-temperature liquid ^4He are closely related phenomena. In the bulk liquid, at saturated vapour pressure, they show up together below the so-called λ temperature ($T_s = T_{\text{BEC}} = T_\lambda = 2.17\text{ K}$).

The superfluid properties (macroscopic in nature) of liquid ^4He confined in several porous media have been extensively investigated [1]. It has been found that confinement reduces the superfluidity temperature T_s . The smaller the pore size of the porous media, the further T_s is reduced below T_λ .

From the microscopical point of view, the existence of p-r excitations is considered a proof of BEC. The first measurement of the characteristic phonon-roton excitations of super ^4He confined in porous media by inelastic neutron scattering was performed by Coddens et al. [2] in 1994.

The initial goal in these and subsequent measurements was to reveal how confinement and disorder modify the phonon-roton excitations from their bulk ^4He values.

Apart from additional weak, relatively broad, side excitations, believed to be supported by denser liquid layers close to substrate surface [3], no major changes have been detected in the microscopical behaviour of the liquid once confined.

1.1. Aim

Two of the possible sources of errors in the above results are:

1. a too large average pore size for the confining medium
2. the presence of bulk liquid helium embedding the porous material and the confined helium sample

A too broad pore size distribution of the confining medium could also non trivially affect results [4].

We will deal with these points and try to overcome them.

2. Materials

Recently discovered materials as silica MCM-41 seem to provide good confining environments: pores are a collection of bunches of cylindrical pores whose average diameter ranges from 20 Å to 100 Å following synthesis procedure, while pore size distribution has a sharp-peaked shape.

3. Tools

In order to characterize the confined ^4He sample we performed (volumetric-)pressure isotherms using ^4He as probe gas. The technique consists in measuring the amount of ^4He adsorbed by the porous sample, N_{ads} , at fixed temperature T vs. equilibrium pressure, P . This quantity is obtained by direct subtraction of the residual gas amount at equilibrium, N_{gas} , from the total injected gas amount, N_{inj} .

It is customary to plot results of such a measurement as specific adsorbed amount of matter, n_{ads} , vs. reduced pressure p ; these two variables are defined

as: $n_{\text{ads}} = \frac{N_{\text{ads}}}{m}$ and $p = \frac{P}{P_0(T)}$, where m is the mass

of the adsorbing sample and $P_0(T)$ is saturated vapour pressure of the probe gas at temperature T .

The measurements of gas amounts are performed by high accuracy (0.12% on reading) pressure measurements over calibrated (about 0.1% accuracy) volumes. Some features can be extracted directly from the adsorption diagram. In particular, a step in filling at nearly constant pressure in the range $0.1 \leq p \leq 0.9$, is generally associated with the condensation of the probe inside pores into a state very similar to bulk condensed phase. This phenomenon is called "capillary condensation" and occurs in the so-called "mesoporous" materials that is to say, in materials presenting pores whose size ranges from 20 Å to 50 Å. The capillary condensed phase is actually the phase we are interested in: a confined liquid phase. Performing experiments below saturated vapour pressure make us sure that no bulk liquid is surrounding the confined sample and that all the observed signal is coming from confined ^4He .

4. Results

A MCM-41 sample has been provided by University of Mulhouse (France); average pore size is $d = (32 \pm 3)\text{ Å}$ and specific surface is $s \cong 910\text{ m}^2\text{g}^{-1}$ as determined by standard nitrogen adsorption measurements and data analysis in the framework of the Barrett, Joyner and Halenda model [5].

We performed ^4He pressure isotherms at $T = (2.48 \pm 0.01)\text{ K}$; results are shown and commented in figure 1; we stress that the sample allows capillary condensation of helium at this temperature. Exploratory neutron scattering experiments have been performed on the MIBEMOL inelastic spectrometer at LLB using an incident neutron wavelength of 5 Å. Results are shown and



commented in figure 2. The main achievement of these measurements is that they show undoubtedly that the inelastic signal generating from confined helium (in the range of exchange energy considered) is mainly due to capillary condensed liquid. This inelastic signal appears to have roughly the same dispersion as expected for the 3D bulk liquid.

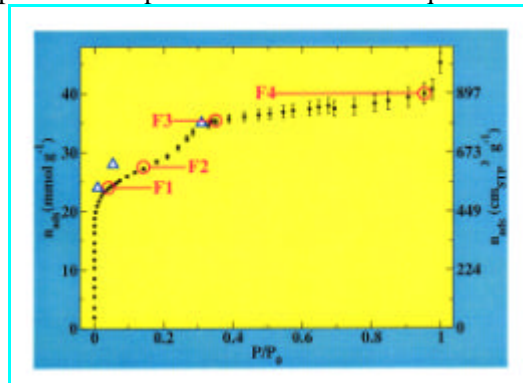


Figure 1. Helium adsorption diagram on a 32 Å pore diameter MCM-41 sample. Black symbols with error: measured isotherm at $T = (2.48 \pm 0.01)$ K; red circles: selected fillings for neutron scattering experiments; blue triangles: values actually obtained at $T = (1.2 \pm 0.1)$ K while performing a neutron scattering experiment on MIBEMOL (LLB). A large amount of helium (up to $F1 \approx 24 \text{ mmol g}^{-1}$) is strongly attached to surface being at equilibrium at a tiny pressure; between F1 and $F2 \approx 28 \text{ mmol g}^{-1}$, some more helium is adsorbed into less favorable adsorption sites, from F2 to $F3 \approx 36 \text{ mmol g}^{-1}$, capillary condensation occurs. At $F4 \approx 40 \text{ mmol g}^{-1}$, the filling is complete and any subsequent added helium amount is condensed into the bulk phase at $P = P_0(T)$ i.e. $p = 1$.

We then performed an experiment on the IN6 spectrometer (ILL, Grenoble) using an incident neutron wave-length of 4.62 Å. We found that excitations of helium confined into MCM-41 show the same features as in other confining materials. In particular energies and lifetimes of the 3D excitations obtained at filling F3 were the same as the bulk liquid within experimental accuracy. Usual side excitations believed to arise from denser liquid layers close to the substrate surface were detected as well.

The original result concerns excitations supported by the layered liquid (filling F2) that disappear at filling F3 (see Fig.3). This excitation is believed to be a

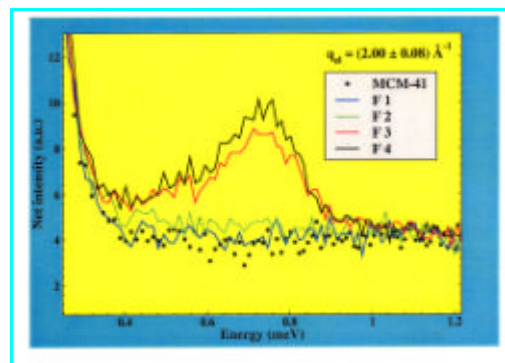


Figure 2. Neutron scattering raw spectra for 32 Å-diameter-pore MCM-41 at selected fillings in helium. Shown spectra have been taken at constant scattering angle; the corresponding value for $q_{el} \equiv q(\omega = 0)$ is $q_{el} = (2.00 \pm 0.08) \text{ Å}^{-1}$. We note that the most of inelastic intensity in the range considered is rising from the capillary condensed phase, despite of its relatively small amount. Some intensity could be associated with filling F2.

capillary wave (also called a ripplon) at the liquid-vapour boundary, supported by liquid layered onto the inner cylindrical surface of the pores. These excitations have been already observed in liquid helium layers on a graphite substrate [6]. Upon complete filling of the pores by capillary condensation, the liquid-vapour interface is strongly depressed and the excitation is no more detected. At filling F3, we start detecting typically 3D excitations instead.

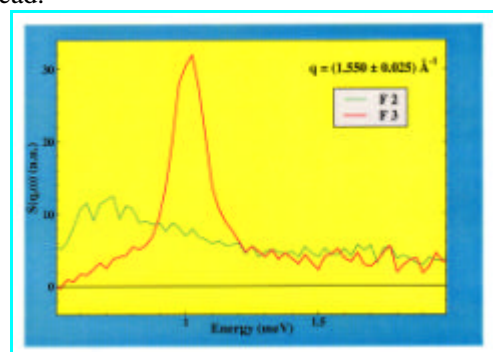


Figure 3. Neutron scattering spectra for 32 Å-diameter-pore MCM-41 at selected fillings in helium. Data have been reduced to constant- q scans after subtracting the MCM-41 contribution. An excitation supported by helium at filling F2 appears to be destroyed upon further filling.

References

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