



Evidence for a Common Physical Origin of the Landau and BEC Theories of Superfluidity

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There are two renowned theories of superfluidity in liquid ^4He , quite different and each with specific domains of application. In the first, the Landau theory, superflow follows from the existence of a well-defined collective mode supported by dense liquid ^4He , the phonon-roton mode. In the second, superflow is a manifestation of Bose-Einstein condensation (BEC) and phase coherence in the liquid. We present combined measurements of superfluidity, BEC and phonon-roton (P-R) modes in liquid ^4He confined in the porous medium MCM-41. The results integrate the two theories by showing that well-defined P-R modes exist where there is BEC. The two are common properties of a Bose condensed liquid and either can be used as a basis of a theory of superfluidity. In addition, the confinement and disorder suppresses the critical temperature for superfluidity, T_c , below that for BEC creating a localized BEC “phase” consisting of islands of BEC and P-R modes. This phase is much like the pseudogap phase in the cuprate superconductors.

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Ever since the initial discovery of superfluidity [1,2] in liquid ^4He in 1938, there has been a rich and vigorous debate on the interdependence of Bose-Einstein condensation (BEC), well-defined collective modes, and superfluidity. For example, immediately following the discovery, London [3] proposed that superflow was a consequence of BEC, the condensation of a macroscopic fraction of the fluid into a single quantum state. In 1941 and 1947, Landau proposed [4,5] that superflow arose because dense liquid ^4He supported a well-defined collective mode, denoted the phonon-roton (P-R) mode. Landau made no reference to BEC. On the basis of P-R modes, Landau constructed a theory of superfluidity so successful that other theories and BEC were largely forgotten. Feynman and Cohen [6] developed a microscopic theory of P-R modes that were eventually observed [7–9]. Modern measurements [10–15] show that in bulk liquid ^4He well-defined P-R modes (see Fig. 1) exist only in the superfluid phase.

BEC, which is more difficult to observe in liquid ^4He , was eventually detected [19] in 1968. Modern measurements [20–23] show that BEC is similarly observed only in the superfluid phase with no BEC in the normal liquid. Modern theories [24–27] generally formulate superfluidity as a consequence of BEC and phase coherence. The superfluid density can be formally related to the condensate. However, even today the explanation of many temperature dependent phenomena relies on the Landau theory.

Superflow of liquid ^4He in porous media has been extensively investigated as an integral part of superfluid studies [28]. Among many fascinating features, the critical temperature for superflow in porous media, T_c , is suppressed below the bulk liquid value, T_λ . The smaller the pore size, the further T_c lies below T_λ . In small pore media T_c goes toward zero at higher pressure [29,30].

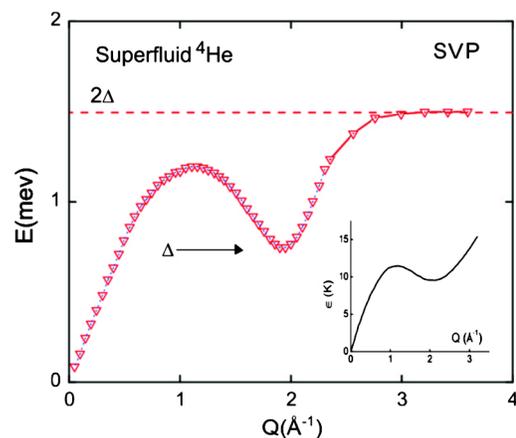


FIG. 1 (color online). The P-R mode energy dispersion curve in superfluid ^4He at low temperature; data are from Andersen *et al.* [16], from Donnelly [17] ($Q \leq 0.4 \text{ \AA}^{-1}$), and from Glyde *et al.* [18] ($Q \geq 2.6 \text{ \AA}^{-1}$). The Δ is the roton energy. The inset shows the P-R mode dispersion curve proposed by Landau [5] in 1947 (1 meV = 11.6 K).

In very small diameter channels, liquid ^4He displays novel two- and even one-dimensional character [31,32].

More recently, the P-R modes of liquid ^4He in several porous media have been measured [33–36]. In contrast to bulk ^4He , well-defined P-R modes are observed at temperatures above T_c . P-R modes are observed up to a temperature denoted T_{PR} . At SVP, $T_{\text{PR}} \approx T_\lambda$ in Vycor [33,37] and gelsils [34,38]. At higher pressure in MCM-41, T_{PR} is consistent with T_λ . The temperature range $T_c < T < T_{\text{PR}}$ has been interpreted [29,33,34,36,37,39] as a “phase” of localized BEC (LBEC).

We present combined measurements of BEC, P-R modes, and superfluidity of liquid ^4He confined in the same porous medium MCM-41 (47 Å diameter pores) where predominantly 3D behavior is anticipated. The measurements of BEC are the first ever in a porous media. The joint measurements of BEC and superflow show unambiguously that the critical temperature for BEC, T_{BEC} , lies above that for superflow, T_c , in MCM-41. The measurements of P-R modes show that well-defined modes are observed up to T_{BEC} , but not above T_{BEC} , i.e. $T_{\text{PR}} = T_{\text{BEC}}$. This experimental result supports theoretical arguments [24,26,40–42] that well-defined modes, especially at wave vectors in the roton region that are essential for the Landau theory of superfluidity, are a consequence of BEC. This finding essentially integrates the two theories of superfluidity: well-defined P-R modes and BEC coincide, with P-R modes a consequence of BEC, and superfluidity can be shown to follow from either.

In addition, the present results also show that in liquid ^4He in porous media (bosons in disorder), there is the temperature range $T_c < T < T_{\text{BEC}}$ in which there is BEC and well-defined P-R modes but no superflow. This supports the earlier interpretation [29,33,34,36,37,39] that $T_c < T < T_{\text{BEC}}$ is a region of localized BEC (BEC localized to patches or islands) arising from disorder. The islands of BEC have independent phases. There is no connected or continuous phase across the liquid as needed for observable superflow. Equally, in the region $T_c < T < T_{\text{BEC}}$, the localization of BEC in space by disorder may be interpreted as leading to phase fluctuations that destroy superflow, as in the pseudogap region of cuprate superconductors. Below we present the measurements of superfluidity, of BEC and of P-R modes in MCM-41.

Superfluid densities, ρ_S , are traditionally measured using torsional oscillators (TO). The oscillator frequency is set by the ratio of a spring constant and the mass of sample in the oscillator. The present sample consists of a powder of MCM-41. In measurements of superflow, it is necessary to have connected liquid across the sample. This requires having liquid ^4He confined in the MCM-41 pores and bulk liquid ^4He lying between the grains of the MCM-41 powder. (This is not the case in the measurements of BEC presented below.) When a fraction of the liquid becomes superfluid and ceases to rotate with the torsional oscillator, the

oscillator frequency, $f(T)$, increases. Figure 2 (top) shows the increase $\Delta f(T) = f(T) - f(T_\lambda)$ in the frequency as the liquid ^4He is cooled. At $T = T_\lambda$, where the bulk liquid ^4He between the grains of MCM-41 becomes superfluid, there is a marked increase in $\Delta f(T)$. At a pressure of 0.1 MPa, $T_\lambda \approx 2.12$ K. When the MCM-41 pores are filled with liquid helium, there is a second marked increase in $\Delta f(T)$ at T_c , the temperature at which the ^4He in the pores becomes superfluid. When there is solid nitrogen (N_2) in the MCM-41 pores, only the bulk ^4He between the grains can become superfluid. In that case $\Delta f(T)$ increases below T_λ only and $\Delta f(T)$ is smaller and proportional to the bulk liquid $\rho_S(T)$. Of the volume open to ^4He in the MCM-41, 30.7% is pore volume and 69.3% is between the grains [43].

Figure 2 (bottom) shows $\Delta f(T)$ of the TO as a function of pressure when there is liquid ^4He both between the grains and confined in the pores. We see that both the critical

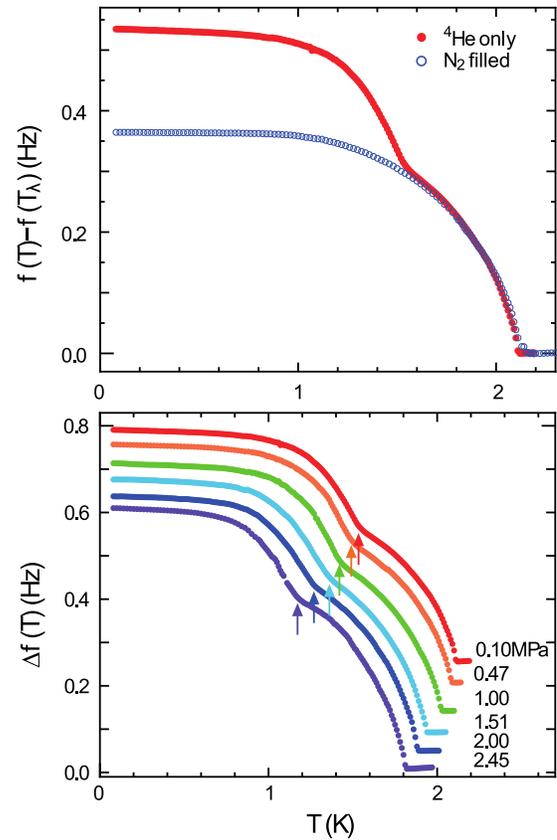


FIG. 2 (color online). Top: change in torsional oscillator frequency $\Delta f(T) = f(T) - f(T_\lambda)$ as the MCM-41 sample is cooled at a pressure of 0.1 MPa below T_λ . There is a marked increase in $f(T)$ at $T_\lambda = 2.12$ K when the bulk liquid between the MCM-41 grains becomes superfluid and a second marked increase at $T_c = 1.58$ K when the liquid ^4He in the MCM-41 pores becomes superfluid. When the MCM-41 pores are filled with nitrogen (N_2), there is helium between the grains only and an increase at T_λ only. Bottom: change in frequency as a function of pressure between 0.10 MPa and 2.45 MPa. The T_c of liquid ^4He in the MCM-41 pores is marked with an arrow.

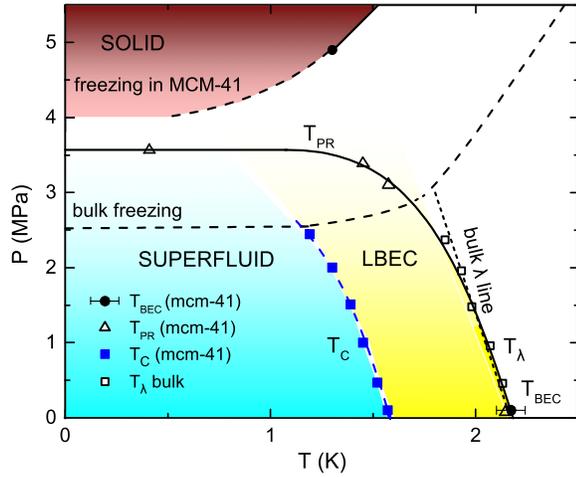


FIG. 3 (color online). Phase diagram of ^4He confined in MCM-41 pores. T_c is the critical temperature for superfluidity of liquid ^4He in the MCM-41 pores. T_λ is the corresponding temperature in bulk liquid ^4He . T_{BEC} is the critical temperature for BEC in the pores at SVP ($p \approx 0$), the same as T_λ . T_{PR} (triangles and solid line) is the temperature at which the intensity in the P-R mode in the pores goes to zero. Within precision $T_{\text{PR}} = T_\lambda$ for $p \leq 2.5$ MPa.

temperature T_λ at which the bulk liquid becomes superfluid and the critical temperature T_c at which the liquid ^4He confined in the pores becomes superfluid decrease with increasing pressure. In this way both T_λ and T_c shown in the phase diagram of Fig. 3 are determined. T_c in MCM-41 pores lies approximately 0.5 K below T_λ .

To date, BEC condensate fractions, n_0 , and P-R modes in liquid ^4He have been measured exclusively using neutron scattering techniques. Using neutrons, BEC and modes in liquid ^4He in the interior of the MCM-41 can be observed directly. To observe liquid ^4He exclusively in the pores, we need liquid in the pores but no bulk liquid between the grains. This requires a pressure below the saturated vapor pressure (SVP) of bulk liquid ^4He . Specifically, helium is highly attracted to the walls of SiO_2 porous media such as MCM-41. The ^4He first added to MCM-41 is deposited at low pressure on the medium walls, up to 22 mmol/gm or approximately 1.5–2 layers of irregular amorphous ^4He solid on the rough pore walls [44]. The subsequent helium added is still attracted to the pores and is deposited as liquid in the interior of the pores. Only after the pores are full does the vapor pressure rise to SVP, the pressure needed for bulk liquid to form. In our measurements of BEC and P-R modes presented below, we added sufficient ^4He to fill the pores but have no bulk liquid between the grains. An adsorption isotherm of ^4He in the present MCM-41 appears in Ref. [44].

In bulk liquid ^4He at low temperature, n_0 ranges [22,45] from $7.25\% \pm 0.75\%$ at SVP to $2.88\% \pm 0.60\%$ at 2.40 MPa. The bulk liquid solidifies at $p = 2.53$ MPa. Figure 4 shows the condensate fraction of ^4He (solid layers plus liquid) in the pores of MCM-41. The observed fraction

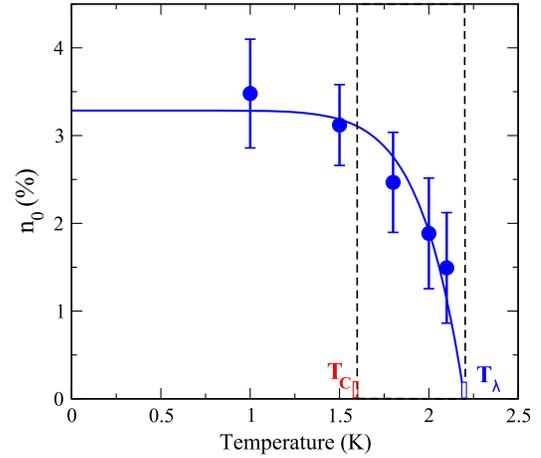


FIG. 4 (color online). Condensate fraction, n_0 , of ^4He in the pores of MCM-41 at SVP. The points with error bars are data and the solid line is a fit to $n_0(T) = n_0(0) [1 - (T/T_\lambda)^\gamma]$ to data with $n_0(0) = 3.3 \pm 0.40\%$ and $\gamma = 9.1$ for $T < T_\lambda$. The errors shown arise from statistical errors in the neutron scattering measurement. The critical temperature for BEC in MCM-41, T_{BEC} , is the same as the bulk value $T_\lambda = 2.17$ K.

shown in Fig. 4 is less than the bulk value ($n_0 = 7.25\%$) because the ^4He in the amorphous solid layers is not expected to support a condensate. Roughly 45% of ^4He is the “inert” solid layers. If we correct for these solid layers, the condensate fraction in the liquid ^4He in the interior of the pores is $n_0 \approx 6\%$, slightly less than the bulk value. The essential result is that T_{BEC} of liquid ^4He in MCM-41 is the same as $T_{\text{BEC}} = T_\lambda$ in bulk liquid ^4He . Also, as we see from Fig. 3, and as discussed in the section below, T_{PR} , the temperature at which the intensity in the P-R mode goes to zero, is the same as T_λ within experimental resolution.

Well-defined phonon-roton modes of liquid ^4He have now been observed in several porous media. Remarkably, in fully filled pores at SVP, the P-R mode energy is the same as that in bulk liquid in all porous media investigated, within precision. Equally remarkably at SVP, well-defined P-R modes are observed at the temperatures above T_c , e.g. in Vycor [33,37] and gelsils [34,38]. While the intensity in the mode decreases with increasing temperature, the mode continues to be observed up to a temperature, T_{PR} , which is approximately equal to the bulk critical temperature, T_λ . In liquid ^4He under pressure in gelsil and MCM-41, P-R modes are also observed above T_c in gelsil. For example, at 3.4 MPa in gelsil where T_c goes to $T = 0$ K [29] $T_{\text{PR}} \approx 1.5$ K [46]. At higher pressure, there is bulk solid between the grains and liquid in the pores. In Fig. 3, the line with triangles marks T_{PR} in the present MCM-41 at higher pressures [36]. These data were interpreted as a signature that there is BEC in porous media above T_c up to temperatures $T_{\text{BEC}} \approx T_\lambda$. The present direct observation of BEC up to T_λ in MCM-41 at SVP shown above in Fig. 4 confirms this interpretation.

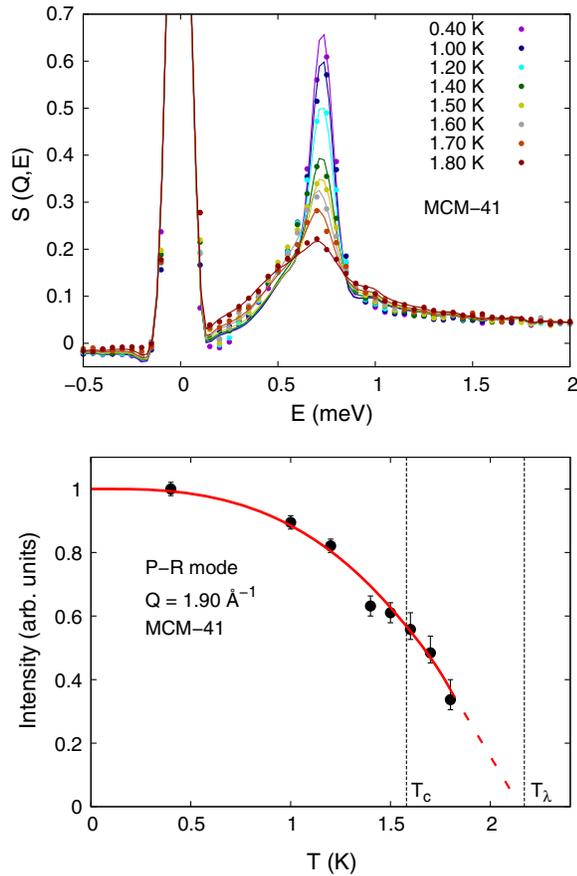


FIG. 5 (color online). Top: net dynamical structure factor, $S(Q, E)$, at $Q = 1.9 \text{ \AA}^{-1}$ as a function of temperature of liquid ^4He and solid layers at SVP in the pores of MCM-41. The large peak at $E = 0$ is elastic scattering. The peak at $E = 0.7 \text{ meV}$ is the roton. A layer mode contributes some broad intensity on the low E side of the roton peak. The intensity in the roton clearly decreases with increasing temperature. Bottom: integrated intensity in the roton peak as a function of temperature. The intensity in the P-R mode extrapolates to zero at a temperature $T_{\text{PR}} \approx T_{\lambda}$.

Figure 5 (top) shows the dynamic structure factor, $S(Q, E)$, of liquid ^4He in the present MCM-41 at SVP at the roton wave vector $Q = 1.9 \text{ \AA}^{-1}$. The large intensity at $E = 0$ is elastic scattering from the MCM-41. The peak at $E = 0.7 \text{ meV}$ is the P-R mode plus a layer mode contributing broad intensity to the lower energy side of the peak. The intensity in the P-R mode decreases with increasing temperature. This intensity is transferred to broad intensity at higher temperature that contributes over a wide energy range attributed to scattering from the normal fluid. Figure 5 (bottom) shows the integrated intensity in the P-R mode as a function of temperature. The intensity in the P-R mode can be obtained by fitting models of $S_{\text{PR}}(Q, E)$ to the data and integrating over E . The intensity as a fraction of the intensity at $T = 0.4 \text{ K}$ is shown. At $T = T_c$, there is clearly a well-defined mode. The intensity extrapolates to zero at $T_{\text{PR}} \approx T_{\lambda}$. Thus in the same MCM-41 at

SVP we have unambiguously shown that $T_{\text{PR}} > T_c$ and $T_{\text{PR}} \approx T_{\text{BEC}} = T_{\lambda}$. The direct observation of P-R modes up to T_{BEC} confirms that P-R modes exist where there is BEC.

We have presented measurements of both the superfluid density, $\rho_s(T)$, and the BEC condensate fraction, $n_0(T)$, of liquid ^4He confined in 47 \AA diameter pores. The confinement to nanocales and the disorder introduced by the porous media suppresses the critical temperature for superfluidity, T_c , below the bulk liquid value, T_{λ} . In contrast, the critical temperature for BEC is unaffected within precision and remains at the bulk liquid value, $T_{\text{BEC}} \approx T_{\lambda}$. In confinement, T_c and T_{BEC} are separated. Also in confinement, well-defined P-R modes are observed up to a temperature denoted T_{PR} . The present measurements show that at SVP $T_{\text{PR}} = T_{\text{BEC}}$. At higher pressure T_{PR} is consistent with T_{BEC} . Well-defined P-R modes at higher wave vector Q (particularly well-defined rotons) are associated with BEC. Phase coherence and well-defined P-R modes both arise from BEC and are two properties among several of a Bose condensed liquid. This effectively reconciles the Landau (P-R mode) and BEC (phase coherence) theories of superfluidity since they are based on consistent properties that have a common origin. They were presented originally as based on quite different physics and representing conflicting views on the origin of superfluidity.

The connection between BEC and well-defined phonon-like modes was first discussed by Bogoliubov [40]. He showed that when there is BEC in a Bose gas, the single particle and density mode have the same energy. This was extended to a dense Bose liquid by Gavoret and Nozières [41] and others [24,26]. When there is BEC, the density and single particle response of a Bose liquid have the same poles (same energy vs Q) at all wave vectors. Thus there is no independent single particle modes at low energy to which the P-R mode can decay, as there are in normal Fermi and Bose systems [47]. The P-R mode can decay only by interaction with other P-R modes and this decay rate is very small at low temperature leading to well-defined P-R modes. The present measurements confirm that well-defined P-R modes coincide with BEC.

To discuss the impact of confinement on the condensate fraction, we note that $n_0 = N_0/N$ at $T = 0 \text{ K}$ in bulk helium depends chiefly on the liquid density. The density may be characterized by the ratio of the volume occupied by the bosons relative to volume of the liquid. Per particle, this ratio is (a^3/v) where a is the Boson hard core diameter and $v = V/N = n^{-1}$ is the volume per Boson. When $na^3 \ll 1$, the gas is dilute, the bosons are little localized in real space, and n_0 can be large; i.e. the bosons can be highly localized in momentum space. When $na^3 \approx 0.2$ as in liquid ^4He , the bosons are highly localized in real space and so cannot be localized in momentum space and n_0 is small. The length scale that sets n_0 is the inter-boson spacing. Thus confinement in a porous medium on a scale 10–20 times the inter-boson spacing affects n_0 little

directly. Indirectly, the ^4He is highly attracted to the pore walls. This increases the density of and solidifies the layers adjacent to the walls reducing n_0 to zero in these layers. However, the ^4He in the interior of the pores remains liquid near or at the bulk density leaving n_0 and T_{BEC} close to the bulk liquid values in the interior of the pores.

In summary, the present direct measurements of BEC and P-R modes show that P-R modes and BEC coexist and confirm the prediction that well-defined modes in liquid ^4He arise because of BEC. The direct measurements of BEC and superfluidity confirm that there is a temperature range where there is BEC but no superflow interpreted as a LBEC region similar to the pseudogap phase.

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