Quantum Critical Phenomena of $^4$He in Nanoporous Media

Thomas Eggel,\textsuperscript{1} Masaki Oshikawa,\textsuperscript{1} and Keiya Shirahama\textsuperscript{2}
\textsuperscript{1}Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581 Japan
\textsuperscript{2}Department of Physics, Keio University, Yokohama 223-8522 Japan
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The superfluid transition in liquid $^4$He filled in Gelsil glass observed in recent experiments is discussed in the framework of quantum critical phenomena. We show that quantum fluctuations of phase are indeed important at the experimentally studied temperature range owing to the small pore size of Gelsil, in contrast to $^4$He filled in previously studied porous media such as Vycor glass. As a consequence of an effective particle-hole symmetry, the quantum critical phenomena of the system are described by the 4D XY universality class, except at very low temperatures. The simple scaling agrees with the experimental data remarkably well.

Introduction– Superfluidity is one of the most impressive macroscopic quantum phenomena that can be experimentally observed. Since liquid $^4$He is a very clean system and the U(1) symmetry of the quantum phase is exact, its superfluid transition is also an ideal case to study phase transitions. In fact, much of the most precise experimental estimates of critical exponents is obtained from the superfluid transition of $^4$He \cite{1,2}.

In bulk liquid $^4$He, the second-order superfluid phase transition occurs at finite temperature only. On the other hand, the properties of liquid $^4$He can be changed by confining $^4$He in porous media. For example, $^4$He in Vycor glass exhibits a quantum phase transition as a function of film thickness, when $^4$He forms a two-dimensional film on the interior surface of the pores \cite{3}. However, the physics might be complicated due to the crossover from two-dimensional to three-dimensional behaviors. When $^4$He is filled in Vycor pores, the superfluid transition remains at finite temperature (see \cite{4} and references therein). Recently, however, by using Gelsil glass which has nanopores of 2.5 nm mean nominal diameter, considerably smaller than comparable substrates used in the past, the superfluid transition temperature is suppressed to zero under an applied pressure \cite{5}. This implies the existence of a quantum critical point at zero temperature.

The non-superfluid phase close to the quantum critical point exhibits rather abnormal properties. This also seems to be closely related to the pseudogap regime in high-$T_c$ superconductivity \cite{6}. The $^4$He in nanoporous media is in several ways (including the absence of fermionic degrees of freedom) simpler, and might provide a useful insight into the physics of more complex systems such as the high-$T_c$ superconductors.

In this Letter, we discuss the quantum critical phenomena of the superfluid transition in liquid $^4$He in Gelsil glass. We determine the value of the interaction strength $V$ in the Bose-Hubbard model for liquid $^4$He in porous media. Although the strongly interacting and dense quantum liquid nature of $^4$He makes a microscopic theoretical calculation of the effective parameter $V$ difficult, we show that it can be derived from macroscopic properties of liquid $^4$He. We find that, although the interaction parameter is about 20 mK for the Vycor system, it is as large as about 1 K for the Gelsil system. This explains the fact that the quantum critical phenomena are visible for the experimentally studied temperature range only in the Gelsil system.

The Bose-Hubbard model with disorder has been a subject of extensive theoretical study. Its quantum critical phenomena are highly nontrivial, and much of the problem still remains open. Nevertheless, we argue that the particle-hole symmetry breaking due to chemical potential, which drives the system away from the 4D XY universality class, is practically negligible in the present system. As a consequence, for a large part of the phase diagram, the quantum critical phenomena observed experimentally in $^4$He in Gelsil glass can be understood with the simple non-random 4 dimensional XY (4D XY) universality class \cite{7}.

We demonstrate that the simple scaling based on the non-random 4D XY universality class indeed agrees quite well with the experimental data on liquid $^4$He in Gelsil glass, except in the small region close to the quantum critical point, where a crossover to a different universality class is observed.

Setup– The Gelsil glass may be modeled by many highly interconnected pores, each of which can contain a number of $^4$He atoms. While the size of the pores is randomly distributed, there is a typical pore diameter for a given sample.

One of the most outstanding experimental findings on the system is a rounded peak in the specific heat at temperature $T_B$ somewhat lower than the superfluid transition temperature $T_\lambda$ in the bulk. In Ref. \cite{8} it was proposed that the specific heat peak should be interpreted as the formation of localized BEC (LBE) within each pore.

In fact, here we demonstrate that the peak temperature $T_B$ can be indeed understood as a rounded lambda transition temperature in a finite-size system. Standard scaling theory gives

$$\Delta T \equiv T_\lambda - T_B(l) \propto C l^{-1/\nu_{3D}},$$

where $l$ is the linear pore size, and $\nu_{3D} \sim 0.67$ is the correlation length exponent for the $\lambda$ transition.

In two samples of nanoporous glasses with different pore sizes, Gelsil and Vycor, different values of $\Delta T$ were observed \cite{5,8}. Moreover, a similar shift was also observed in porous Gold samples with two different pore sizes \cite{9}. The four values of $\Delta T$ are plotted as a function of the pore size $l$ in Fig. \cite{10}. Here we use the effective pore size, obtained by subtracting the inert layer thickness from the nominal pore diam.
eter. Although the data were taken for different porous materials at different pressures, the agreement with the scaling is remarkable. \( \nu_{4D} = 1/1.48 = 0.676 \) estimated from the fit is consistent with the known value \( \nu_{4D} \approx 0.67 \). In addition to establishing the localized BEC picture, this result implies that liquid \(^4\)He in the nanopores does inherit bulk properties.

**Quantum Fluctuations**—It is important to estimate the typical value of the “charging energy” \( V_i \). In the application of the Bose-Hubbard model to superconductors, \( V_i \) represents the charging energy due to Coulomb repulsion. In contrast, in the present case of neutral liquid \(^4\)He, there is no Coulomb repulsion. Nevertheless, putting an extra atom to, or removing an atom from, a pore in the groundstate should lead to an increase in energy. This can be related to the finite compressibility of the liquid in the pore. In fact, we can estimate \( V_i \) as follows:

\[
\frac{1}{V_i} \partial_n \mu_i = V_i \nu^2 \kappa, \tag{4}
\]

where \( V_i \) is the (effective) volume of the pore, and \( \nu \) and \( \kappa \) are the number density and the compressibility of liquid \(^4\)He, respectively. We approximate \( \nu \) and \( \kappa \) of \(^4\)He inside the pores by their values in the bulk: \( \nu \approx 2.1 \times 10^{28} \text{ m}^{-3} \), \( \kappa \approx 10^{-7} \text{ Pa}^{-1} \).

Then, assuming that a typical pore is a sphere with effective diameter \( 2R_{\text{eff}} = 1.3 \) nm (subtracting the thickness 0.6 nm of the inert layer), we obtain an estimate for the typical value of \( V_i \) \( \approx 1.4K \) for \(^4\)He in Gelsil Glass. In contrast, for Vycor Glass with effective diameter 5 nm, the same argument leads to \( V_i \approx 0.02K \).

If \( T \gg V_i \), the quantum fluctuations of the phase may be neglected. In such a limit, although the phase \( \theta_i \) is a quantum-mechanical degree of freedom, it can be regarded as a classical variable. On the other hand, when \( T \lesssim V_i \), quantum fluctuations of the phase become important. Thus, in the temperature range probed in the experiments, quantum fluctuations would be important for \(^4\)He filled in Gelsil glass but not in Vycor glass. This is consistent with the experimental results that quantum critical phenomena are observed only in Gelsil glass, when pores are filled by \(^4\)He.

In the continuum limit, the (disordered) Bose-Hubbard model is described by the Lagrangian density in \( 3 + 1 \) dimensions:

\[
\mathcal{L} = \frac{1}{2} |\nabla \psi|^2 - \frac{1}{2} \psi^* \nabla \cdot (g_0 - \delta g(x)) |\psi|^2 + \frac{1}{2} (r_0 + \delta r(x)) |\psi|^2 + \frac{1}{4} u_0 \psi^4, \tag{5}
\]

where \( \psi \) is a complex scalar field, \( g_0, r_0, \) and \( u_0 \) are constants, and \( \delta g(x) \) and \( \delta r(x) \) are position-dependent random variables distributed around zero.

When \( g_0 = \delta g = \delta r = 0 \), this theory reduces to the standard \( \psi^4 \) theory with the dynamical critical exponent \( z = 1 \), and the quantum critical point is described by the 4D XY universality class. Since \( 4 \) is the upper critical dimension for the XY model, the critical exponents are given by the mean-field theory. For example, the correlation length exponent is given by \( \nu_{4D} = 1/2 \). This limit corresponds to, in terms of the original model, the special case where there is no disorder and \( \bar{\delta} \)’s are exactly an integer. In particular, \( g_0 = \delta g = 0 \) implies an exact particle-hole symmetry.

In reality, \( \bar{\delta} \) is randomly distributed, breaking the particle-hole symmetry. Its effect can be classified into the overall
symmetry breaking $g_0$ and the local symmetry breaking $\delta g$, in the continuum theory [5]. $g_0$ is generically non-vanishing and is a relevant perturbation to the 4D XY fixed point. In fact, even in a system without randomness, $g_0$ drives the system to a different critical behavior except at special multicritical points (tips of the Mott lobes). Nevertheless, here we argue that the effect of $g_0$ is practically negligible for the present system, partly owing to the randomness.

Let us assume that the distribution of the pore radius has standard deviation of, say, $\Delta R = 0.02\text{nm}$, which would be rather an underestimation. This translates to the width

$$\Delta \bar{n} \sim 4\pi DR_{\text{eff}}^2 \Delta R \sim 2.2$$

for the distribution of the $\bar{n}_i$. We assume that the $\bar{n}_i$ follow a Gaussian distribution with average $\bar{n}_{av}$ and standard deviation $\Delta \bar{n}$. Since the effect of the particle-hole symmetry breaking is a periodic function of $\bar{n}_i$, it may be estimated by $\sin 2\pi \bar{n}_i$.

The effective overall particle-hole symmetry breaking $g_0$ then reads

$$\int \sin (2\pi x) e^{-\frac{(x-\bar{n}_{av})^2}{2\pi (\Delta \bar{n})^2}} dx = \sin (2\pi \bar{n}_{av}) e^{-2\pi^2 (\Delta \bar{n})^2}.$$ (7)

The first factor $\sin (2\pi \bar{n}_{av})$ just represents the particle-hole symmetry breaking for the average value $\bar{n}_{av}$, which is generically non-vanishing. The second factor $e^{-2\pi^2 (\Delta \bar{n})^2}$ shows the suppression of the symmetry breaking by the random distribution.

For the width given in eq. (6), the suppression is in fact about $10^{-43}$. Thus, in the realistic temperature range, the overall particle-hole symmetry breaking $g_0$ is negligible. That is, in the present system, each pore contains enough particles so that the distinction between the particle and hole becomes unimportant. A similar discussion was given for the asymptotic low-energy behavior of disordered bosons [11]. Here the situation is somewhat different in that the smallness of $g_0$ is not a consequence of the RG transformation, but is rather of microscopic origin.

The random particle-hole symmetry breaking $\delta g$ is an irrelevant perturbation to the 4D XY (Gaussian) fixed point [11]. Although the presence of $\delta g$ is believed to be important to determine the eventual fate of the RG flow, it can be ignored in the neighborhood of the 4D XY fixed point. Thus $\delta R$ and $\delta r$ are the only remaining relevant parameters around the 4D XY fixed point. This theory represents the critical behavior of a classical XY model in 4 dimensions. In the absence of the random $\delta r$, the quantum phase transition belongs to the 4D XY universality class. $\delta R$ corresponds to the temperature of the classical XY model, and is the control parameter for the quantum phase transition. It is thus identified with the pressure $p$ of $^4\text{He}$ in the system.

The random term $\delta r$ corresponds to disorder in the classical XY model. We assume that the disorder is spatially uncorrelated. However, in the mapping to the classical XY model, the disorder is completely correlated in the imaginary time direction. This class of disorder turns out to be a relevant perturbation to the 4D XY fixed point, driving the system to a different fixed point [13]. This is in contrast to the effect of disorder on the finite temperature superfluid transition, where the uncorrelated disorder is an irrelevant perturbation according to the Harris’ criterion [14][15].

The new fixed point induced by $\delta r$, corresponding to the four-dimensional XY model with disorder correlated in the imaginary time direction, is sometimes called the random rod fixed point. At this random rod fixed point, the random part of the chemical potential $\delta \mu$ is believed to be relevant, eventually driving the system to yet another (disordered boson) fixed point [11]. However, if $\delta r$ is small, the system may be described by the 4D XY fixed point and the crossover away from it due to $\delta r$, down to a certain temperature.

We argue that this is indeed the case in the present system of $^4\text{He}$ in Gelsil glass, concerning the experimentally studied temperature range. In fact, as we will demonstrate in the following, a large part of the experimental data fits quite well with the simple 4D XY scaling. At very low temperatures, we see a crossover from the 4D XY behavior, which would be primarily due to the $\delta r$ perturbation.

![FIG. 2. (color online) Frequency shift in the torsional oscillator experiment, which is proportional to the superfluid density, extrapolated to zero temperature and as a function of the pressure. The experimental data, shown in squares, are taken from Ref [16]. The solid line is the 4D XY linear scaling (8). The agreement between the data and the scaling is remarkable except in the close vicinity of the quantum critical point.](image)
XY universality class. For example, if we had $z = 2$ instead of $z = 1$, it would follow $\rho_\alpha \propto (p_c(0) - p)^{3/2}$ which is clearly inconsistent with the data.

We note that the extrapolation of the linear scaling to zero temperature gives $p_c(0) \sim 3.0$ MPa instead of the 3.4 MPa which is the critical pressure directly obtained from the experimental data. We suppose that the non-random 4D XY scaling is valid for the effective critical pressure $p_{c}^\text{eff}(0) \sim 3.0$ MPa; this is different from the “true” critical pressure $p_c(0)$, which is likely to be affected by the randomness.

Finite temperatures—At finite temperature $T$, the length of the system in imaginary time direction takes the finite value $1/T$ and we impose periodic boundary conditions in this direction, see for example [17]. The finite temperature problem is thus equivalent to the classical XY model on a hyperstrip geometry where the base is an infinite three dimensional simple cubic lattice and the slab width is given by $1/T$. The system at finite temperature is thus amenable to a finite size scaling analysis [13] in the finite imaginary time direction. This allows us to make several predictions on the temperature dependence of the system.

For example, the critical pressure $p_c(T)$ at temperature $T$ follows

$$p_{c}^\text{eff}(0) - p_c(T) \propto T^{1/\nu_{4D}} = T^2. \quad (9)$$

In Fig. 3 we fitted a power-law to the experimental data of the phase boundary between the superfluid and the non-superfluid (LBEC) phases presented in [16]. We find an exponent of 2.13 which is in good agreement with equation (9), while $p_{c}^\text{eff}(0) \sim 3.2$ MPa is slightly larger than that obtained in Fig. 2.

Discussion—We have demonstrated that the experimental data on $^4\text{He}$ in Gelsil glass agrees with 4D XY scaling rather well. This corresponds to the theory [5] with a small perturbation $\delta r$, displaying clean 4D XY quantum critical behavior sufficiently far away from the quantum critical point. However, close to the quantum critical point, the effect of $\delta r$ is enhanced and crossover to a different universality class should occur. In fact, by inspection of Fig. 3 the phase boundary at very low temperatures starts to deviate in a pronounced way from the 4D XY scaling [9]. We also find that, in Fig. 2, the superfluid density at zero temperature also deviates from the 4D XY scaling [8], close to the critical pressure $p_c(0)$.

These effects of the disorder are the subject of future investigations.

We also note that, close to the quantum critical point, we find that the extraction of physical quantities (such as the superfluid density and the critical temperature) from the raw frequency data also becomes rather subtle. A more precise analysis would require a more sophisticated analysis of the raw data based on a better theoretical understanding. In any case, we believe that the analyses presented in this Letter demonstrate the basic validity of the proposed picture.

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