

G.Biroli¹ · F.Zamponi²

A tentative replica theory of glassy Helium 4

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Abstract We develop a quantum replica method for interacting particle systems and use it to estimate the location of the glass transition line in Helium 4. Although we do not fully succeed in taking into account all quantum effects, we make a thorough semiclassical analysis. We confirm previous suggestions that quantum fluctuations promote the formation of the glass and give a quantitative estimate of this effect at high density. Finally, we discuss the difficulties that are met when one tries to extend the calculation to the region of low densities and low temperatures, where quantum effects are strong and the semiclassical expansion breaks down.

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1 Introduction

A series of recent experiments on solid He⁴, initiated by Kim and Chan [1, 2] and then performed by several groups [3], raised, among many others, the important question of whether disorder and fast quenches can induce the formation of a quantum glass phase in Helium 4 and to what extent this new phase is related to the super-solid behavior observed experimentally. Following early numerical and theoretical analyses, that showed that perfect crystals do not support super-solidity [4, 5], and a numerical analysis that proposed the presence of a super-glass phase at intermediate densities [6], Ritner and Reppy [7, 8] showed that fast quenches produce disordered samples with a change in the moment of inertia that corresponds to an extremely high superfluid fraction, of the order of 20%. Since then, whether this behavior can be attributed to the presence of a quantum (super-)glass phase has been the subject of some debate [9, 10, 11, 12, 13, 14, 15].

1: Institut de Physique Théorique (IPhT), CEA, and CNRS URA 2306, F-91191 Gif-sur-Yvette, France

2: Laboratoire de Physique Théorique, UMR 8549, CNRS and Ecole Normale Supérieure, 24 Rue Lhomond, 75231 Paris Cedex 05, France

However, before addressing the difficult question of whether supersolidity is possible, present and possibly enhanced in quantum glasses, it would be desirable to have a full understanding of the nature of the glass transition in presence of strong quantum fluctuations, a subject which is instead very poorly understood.

Indeed, several authors attempted to build a theory of the quantum glass transition [16, 17, 18, 19, 20, 21], but none has so far been able to obtain quantitative results for realistic systems such as Helium 4. One of the main difficulties is that new tools are needed to treat glassiness for quantum interacting particles systems in a consistent way (the majority of previous works focused on spin systems and coarse-grained field theories). Recent mean field studies based on Quantum Mode Coupling Theory (QMCT) [22] and on lattice models [23] gave new insights, and a coherent qualitative picture of the glass transition in quantum hard spheres has now emerged [23, 24]. The result is somehow surprising. Basic intuition suggests that increasing the strength of quantum fluctuations should enable atoms to better explore the phase space through tunneling, and thereby inhibit the onset of dynamical arrest that is necessary for glass formation. In contrast, QMCT predicts [22] that increasing quantum fluctuations, by augmenting the thermal wavelength of the particles (e.g. by reducing their mass), actually favors the glass phase. Indeed, one finds that the glass phase emerges at lower density. This result has also been obtained in the exact solution of mean field quantum lattice glass models [23], and is corroborated by path integral numerical simulations [22]. The analysis of a toy model provided the following intuitive explanation [25]: The classical phase space of a dense particle system is split into many different basins; in the liquid phase the system visits a lot of basins that are not very efficiently packed and therefore have a smaller free volume (the volume accessible to each particle to vibrate around its equilibrium position). In the glass phase, the system instead visits a few rare basins that are very efficiently packed and therefore have a larger free volume. Adding quantum fluctuations, particles can lower their kinetic energy by delocalizing, therefore the kinetic energy gain is proportional to free volume: hence, glassy states have lower kinetic energy and are favored by quantum fluctuations. The net result is that the glass transition line is re-entrant, moving at lower density on increasing quantum fluctuations [22]. For strongly quantum systems, however, the transition changes nature and becomes a first order phase transition between a superfluid phase and the glass [23]. This first order transition is accompanied by phase coexistence, which leads to an heterogeneous “superglass”, where regions of low-density superfluid liquid coexist with regions of a high density glass. See [23, 24] for a more detailed discussion.

The results mentioned above have been obtained on simplified models, therefore they only provide qualitative guideline for the physical behavior of Helium 4. Quantitative analytic calculations of the phase boundaries for systems such as Helium 4 are still missing. Unfortunately, Mode-Coupling Theory is of limited use since it is known to give poor estimates of the transition point and, at least at the present stage, neglects exchange effects [22]. The aim of this paper is to adapt the replica theory of glasses [26, 27, 28, 29, 30], which has been shown to give good quantitative estimates of the glass transition point for classical liquids, to the quantum case; and then apply it to Helium 4.

The paper is organized as follows. First, in section 2, we use the standard prescription of replica theory to derive an expression of the glass free energy in the

semiclassical region. We show that this is enough to obtain a quantitative estimate of the re-entrance effect at high density. In section 3 we speculate on the possible phase diagram of Helium 4 in the high density amorphous metastable region. Next, in section 4, we discuss the assumptions we made and identify the main problems of the theory: we show that there are some important conceptual difficulties that prevent us from extending the calculation to the strongly quantum regime and from including exchange. While we believe that these difficulties can be in principle overcome, we leave this task for future work.

2 The semiclassical replica method

We start our discussion by a straightforward application of the classical prescription of the replica method to the quantum case. We consider a d -dimensional system of quantum Bosonic particles, of mass M , interacting via a potential $v(r)$. As usual h is the Planck's constant and $\hbar = h/2\pi$. We set the Boltzmann constant $k_B = 1$. If x_i is the position of particle i , with $i = 1, \dots, N$, the Hamiltonian is

$$H = -\frac{\hbar^2}{2M} \sum_i \nabla_{x_i}^2 + \sum_{i < j} v(x_i - x_j), \quad (1)$$

where ∇_{x_i} is the gradient with respect to x_i .

In the following we will make an extensive use of the replica theory of classical glasses. We refer to the original works [26, 28] and the review [29] for details. The prescription of the theory is to consider a system made of m copies of the original system, subjected to an inter-replica attractive coupling that we choose to be harmonic [28]. We denote by x_{ai} , $a = 1, \dots, m$, the copies of particle i . The Hamiltonian of such replicated system is

$$H = -\frac{\hbar^2}{2M} \sum_{ai} \nabla_{x_{ai}}^2 + \sum_{a, i < j} v(x_{ai} - x_{aj}) + \frac{\varepsilon}{2m} \sum_{i, a < b} (x_{ai} - x_{bi})^2, \quad (2)$$

where, compared to [28], ε has been rescaled by m for later convenience. Since we want to investigate the semiclassical limit, we neglect exchange for the moment, and consider the particles as distinguishable.

2.1 Small cage expansion

For each molecule i (we now omit the index i) we consider the column vector x such that $x^T = (x_1, \dots, x_m)$. We perform the change of variables $y_a = v_a^T x$ with $v_1^T = \frac{1}{\sqrt{m}}(1, \dots, 1)$, $v_a^T v_1 = 0$ and $v_a^T v_b = \delta_{ab}$. The Jacobian of the transformation is 1 because the vectors v_a are orthonormal. Thus the kinetic term is invariant under the transformation, $\sum_a \nabla_{x_a}^2 = \sum_a \nabla_{y_a}^2$. Defining the matrix of all unit entries, $1_{ab} = 1$, denoting by δ the identity matrix, and using the completeness relation $\delta = \sum_a v_a v_a^T$, we can write $\sum_{a < b} (x_a - x_b)^2 = x^T (m\delta - 1)x = \sum_{ab} x^T v_a v_a^T (m\delta - 1) v_b v_b^T x = \sum_{ab} y_a v_a^T (m\delta - 1) v_b y_b$. It is easy to show that $(m\delta - 1)v_1 = 0$ and

that $(m\delta - 1)v_a = mv_a$ for $a > 1$. Then, $\sum_{a < b} (x_a - x_b)^2 = \sum_{a=2}^m my_a^2$. Thus the Hamiltonian in the y variables reads

$$H = -\frac{\hbar^2}{2M} \sum_{ai} \nabla_{y_{ai}}^2 + \sum_{a,i < j} v(x_{ai} - x_{aj}) + \frac{\varepsilon}{2} \sum_{i,a > 1} y_{ai}^2, \quad (3)$$

where for each molecule i

$$x_a = \sum_b (v_b)_a y_b = \frac{y_1}{\sqrt{m}} + \sum_{b > 1} (v_b)_a y_b = X + \sum_{b > 1} (v_b)_a y_b, \quad (4)$$

with $X = y_1/\sqrt{m} = (\sum_a x_a)/m$ the center of mass of the molecule.

Now, following [28], we perform a large ε expansion. For large ε we assume that the y_a , $a > 1$ are small due to the harmonic term, and we expand the potential $v(x_{ai} - x_{aj})$. For a given pair of indices i, j (that we now omit), we define $\Delta x_a = x_{ai} - x_{aj}$, and it follows from Eq. (4) that $\Delta x_a = \Delta X + \sum_{b > 1} (v_b)_a \Delta y_b$. We denote by $\mu, \nu = 1, \dots, d$ the spatial indices, and $\partial_\mu v = dv/dx^\mu$ the derivative with respect to coordinate x^μ . Then we can easily expand the potential as follows:

$$\begin{aligned} \sum_a v(\Delta x_a) &= mv(\Delta X) + \sum_a \partial_\mu v(\Delta X) \sum_{b > 1} (v_b)_a \Delta y_b^\mu \\ &+ \frac{1}{2} \sum_a \partial_{\mu\nu} v(\Delta X) \sum_{b > 1} (v_b)_a \Delta y_b^\mu \sum_{c > 1} (v_c)_a \Delta y_c^\nu. \end{aligned} \quad (5)$$

Recalling that for $b > 1$, $\sum_a (v_b)_a = \sqrt{mv_1^T} v_b = 0$ and that $\sum_a (v_b)_a (v_c)_a = v_b^T v_c = \delta_{bc}$, we get $\sum_a v(\Delta x_a) = mv(\Delta X) + \frac{1}{2} \partial_{\mu\nu} v(\Delta X) \sum_{b > 1} \Delta y_b^\mu \Delta y_b^\nu$. Collecting the terms together and changing variables to $X_i = y_{1i}/\sqrt{m}$, we finally get

$$\begin{aligned} H &= \underbrace{-\frac{\hbar^2}{2mM} \sum_i \nabla_{X_i}^2 + \sum_{i < j} mv(X_i - X_j)}_{H_0(X)} + \sum_{i,a > 1, \mu} \underbrace{\left[-\frac{\hbar^2}{2M} \nabla_{y_{ai}^\mu}^2 + \frac{\varepsilon}{2} (y_{ai}^\mu)^2 \right]}_{H_{\text{harm}}(y_{ai}^\mu)} \\ &+ \underbrace{\frac{1}{2} \sum_{i < j} \partial_{\mu\nu} v(X_i - X_j) \sum_{a > 1} (y_i - y_j)_a^\mu (y_i - y_j)_a^\nu}_{V_{\text{int}}(X,y)}. \end{aligned} \quad (6)$$

At this order, the Hamiltonian is the sum of a term $H_0(X)$ that describes the system of the centers of mass, with a renormalized mass mM and renormalized interaction $mv(r)$, plus a sum of independent terms $H_{\text{harm}}(y)$, that describe harmonic oscillators of spring constant ε . The centers of mass are coupled to vibrations by the term V_{int} ; since the vibrations are small for large ε , this coupling is small and we can treat it as a perturbation.

2.2 Computation of the partition function

We now want to compute the partition function of the system at temperature T , using the Hamiltonian in (6). Again, we stress that in the following we consider that the particles are distinguishable. Otherwise, even in the absence of the coupling V_{int} , one should consider the y variables as “internal” degrees of freedom (like spins) of the centers of mass, and one should therefore impose the permutation symmetry on the global wavefunction, that would correlate the X and y parts of the wavefunctions. Instead, for distinguishable particles, there is no special symmetry requirement and we consider that in absence of V_{int} the space of wavefunctions is a product of independent wavefunctions for the X and y parts. Under these assumptions, we get, developing at first order in V_{int} :

$$Z_m(\varepsilon) = \text{Tr} e^{-\beta H} = \text{Tr}_X e^{-\beta H_0(X)} \left(\text{Tr}_y e^{-\beta H_{\text{harm}}(y)} \right)^{Nd(m-1)} \left[1 - \beta \langle V_{\text{int}} \rangle + O(V_{\text{int}}^2) \right],$$

where $\langle V_{\text{int}} \rangle$ is the quantum and thermal average of the perturbation over the X and y . The partition function and free energy of each independent oscillator is

$$\begin{aligned} Z_{\text{harm}}(\varepsilon) &= \text{Tr}_y e^{-\beta H_{\text{harm}}(y)} = \frac{e^{\beta \hbar \omega / 2}}{e^{\beta \hbar \omega} - 1}, \\ F_{\text{harm}}(\varepsilon) &= -T \log Z_{\text{harm}}(\varepsilon) = -T \log \frac{e^{\beta \hbar \omega / 2}}{e^{\beta \hbar \omega} - 1} = U_{\text{harm}}(\varepsilon) - TS_{\text{harm}}(\varepsilon), \end{aligned} \quad (7)$$

where the frequency $\omega = \sqrt{\varepsilon/M}$; there are $d(m-1)$ independent oscillators per molecule. It will also be useful to define the mean square displacement of the oscillator:

$$\langle y^2 \rangle_\varepsilon = 2 \frac{dF_{\text{harm}}(\varepsilon)}{d\varepsilon} = \frac{\hbar \omega}{\varepsilon} \left[\frac{1}{2} + \frac{1}{e^{\beta \hbar \omega} - 1} \right] = \frac{\hbar \omega}{\varepsilon} \bar{n}(\omega), \quad (8)$$

with $\bar{n}(\omega) = 1/2 + 1/(e^{\beta \hbar \omega} - 1)$.

The average of the interaction term over y is

$$\begin{aligned} \langle V_{\text{int}} \rangle_y &= \frac{1}{2} \sum_{i < j} \partial_{\mu\nu} v(X_i - X_j) \sum_{a > 1} \langle (y_i - y_j)_a^\mu (y_i - y_j)_a^\nu \rangle_y \\ &= (m-1) \langle y^2 \rangle_\varepsilon \sum_{i < j} \Delta v(X_i - X_j), \end{aligned} \quad (9)$$

using that different spatial components and sites i and j are uncorrelated, $\langle (y_i - y_j)_a^\mu (y_i - y_j)_a^\nu \rangle_y = \delta_{\mu\nu} \langle (y_{ia}^\mu)^2 + (y_{ja}^\mu)^2 \rangle_y = 2\delta_{\mu\nu} \langle y^2 \rangle_\varepsilon$. Taking the average of V_{int} also on the centers of mass, we get the first order correction to the free energy:

$$\begin{aligned} \Phi(m, \varepsilon) &= -\frac{T}{N} \log Z_m(\varepsilon) = -Td(m-1) \log \left[\frac{e^{\beta \hbar \omega / 2}}{e^{\beta \hbar \omega} - 1} \right] \\ &\quad - \frac{T}{N} \log \text{Tr}_X e^{-\beta H_0(X)} + (m-1) \langle y^2 \rangle_\varepsilon \left\langle \frac{1}{N} \sum_{i < j} \Delta v(X_i - X_j) \right\rangle_0 \\ &= d(m-1) F_{\text{harm}}(\varepsilon) + F_0(m) + (m-1) \langle y^2 \rangle_\varepsilon \frac{\rho}{2} \int dr \Delta v(r) g_0(r), \end{aligned} \quad (10)$$

where $F_0(m) = -T/N \log \text{Tr}_X e^{-\beta H_0(X)}$ is the free energy of the effective liquid of the centers of mass, and $\rho^2 g_0(X - Y) = \langle \sum_{i \neq j} \delta(X - X_i) \delta(Y - Y_j) \rangle_0$ is its pair correlation function. In the following we will use the notation $\langle \Delta v(r) \rangle = \frac{\rho}{2} \int dr \Delta v(r) g_0(r)$.

Note that the Hamiltonian $H_0(X)$ can be rewritten as $H_0(X) = m \left[-\sum_i \frac{\hbar^2}{2m^2 M} \nabla_{X_i}^2 + \sum_{i < j} v(X_i - X_j) \right]$, then the fluid of centers of mass can be viewed as the original fluid at inverse temperature $\tilde{\beta} = m\beta$ and with a mass $\tilde{M} = m^2 M$. Therefore:

$$\frac{1}{m} F_0(m) = -\frac{T}{mN} \log \text{Tr}_X e^{-\beta H_0(X)} = F_{\text{liq}}(m\beta, m^2 M), \quad (11)$$

where $F_{\text{liq}}(\beta, M)$ is the free energy associated to the original Hamiltonian (1).

2.3 The replicated free energy

Recalling that $\frac{dF_{\text{harm}}(\varepsilon)}{d\varepsilon} = \frac{1}{2} \langle y^2 \rangle_\varepsilon$, we rewrite (10) as

$$\begin{aligned} \Phi(m, \varepsilon) &= mF_{\text{liq}}(m\beta, m^2 M) + d(m-1) \left[F_{\text{harm}}(\varepsilon) + \frac{dF_{\text{harm}}(\varepsilon)}{d\varepsilon} \frac{2\langle \Delta v(r) \rangle}{d} \right] \\ &\sim mF_{\text{liq}}(m\beta, m^2 M) + d(m-1)F_{\text{harm}}(\varepsilon + \kappa), \end{aligned} \quad (12)$$

having defined the effective spring constant $\kappa = 2\langle \Delta v(r) \rangle/d$. This is correct as long as $\kappa \ll \varepsilon$, which is indeed the case since we are working in the limit of large ε . Now, following [28], we have to extrapolate the free energy obtained in the large ε expansion to the limit of zero coupling ε . This can be done via a Legendre transformation with respect to ε , following closely the classical derivation of [28]. This procedure provides a formally correct treatment that shows that the large ε expansion is equivalent to a small cage expansion [28]. However, we see that in Eq. (12) can set directly $\varepsilon = 0$ and we obtain a meaningful result for the replicated free energy, which indeed coincides with the one obtained via the Legendre transform procedure. For the sake of conciseness, we adopt this procedure here: see [28] for further details on the small cage expansion procedure. Finally, we get for the replicated free energy:

$$\Phi(m) = mF_{\text{liq}}(m\beta, m^2 M) + d(m-1)F_{\text{harm}}(\kappa), \quad (13)$$

where the function $F_{\text{harm}}(\varepsilon)$ is defined in (7) and $\kappa = 2\langle \Delta v(r) \rangle_{\text{liq}}/d$, where the average is over the liquid with temperature T/m and mass $m^2 M$.

Given $\Phi(m)$, the thermodynamic properties of the glass, i.e. the internal energy $f(\beta, m)$ and the complexity $\Sigma(\beta, m)$, are obtained following [26, 28]:

$$f(\beta, m) = \frac{\partial \Phi(m)}{\partial m}, \quad \Sigma(\beta, m) = \beta m^2 \frac{\partial \Phi(m)/m}{\partial m}. \quad (14)$$

Using the relations $S_{\text{liq}}(T) = -\partial_T F_{\text{liq}}(T, M)$ (entropy), $K_{\text{liq}}(T) = -M \partial_M F_{\text{liq}}(T, M)$ (kinetic energy), we get for the equilibrium complexity (i.e. the complexity at $m = 1$):

$$\begin{aligned} \Sigma_{\text{cq}}(T) &= \Sigma(\beta, 1) = S_{\text{liq}}(T) - 2\beta K_{\text{liq}}(T) + \beta d F_{\text{harm}}(\kappa) \\ &= S_{\text{liq}}(T) - d S_{\text{harm}}(\kappa) - \beta [2K_{\text{liq}}(T) - d U_{\text{harm}}(\kappa)]. \end{aligned} \quad (15)$$

In the classical case the last term vanishes due to equipartition, since $U_{\text{harm}} = T$ and $K_{\text{liq}} = dT/2$. This is not *a priori* true in the quantum regime, but it should be true if we assume that the liquid is a superposition of “glassy” metastable states corresponding to harmonic vibrations around amorphous positions. We will comment later on this crucial issue.

2.4 The classical limit

Let us first consider the classical limit $\hbar \rightarrow 0$. We have $\bar{n}(\omega) \rightarrow (\beta\hbar\omega)^{-1}$, and

$$F_{\text{harm}}(\varepsilon) \rightarrow T \log \beta\hbar\omega = T \log \Lambda + \frac{T}{2} \log \frac{\varepsilon}{2\pi T}, \quad (16)$$

where $\Lambda = h/\sqrt{2\pi MT}$ is the thermal wavelength. Moreover the liquid of the centers of mass is classical and has a partition function

$$Z_{\text{liq}} = \int \frac{d^N P d^N X}{h^N N!} e^{-\frac{\beta}{m} \sum_i \frac{p_i^2}{2M} - \beta m \sum_{i<j} v(X_i - X_j)} = \Lambda^{-dN} m^{dN/2} Z_c(\beta m), \quad (17)$$

where $Z_c(\beta) = (N!)^{-1} \int d^N X e^{-\beta \sum_{i<j} v(X_i - X_j)}$ is the configurational partition function. Therefore, $F_0(m) = m F_{\text{liq}}(m\beta, m^2 M) = dT \log \Lambda - \frac{dT}{2} \log m + m F_c(\beta m)$, where $F_c(\beta) = -(T/N) \log Z_c(\beta)$. Using these results, Eqs. (13) and (15) become

$$\begin{aligned} \Phi(m) &= dm T \log \Lambda - \frac{dT}{2} \log m + m F_c(\beta m) + \frac{d(m-1)T}{2} \log \frac{\langle \Delta v(r) \rangle^*}{\pi T d}, \\ \Sigma_{\text{eq}}(T) &= S_{\text{liq}}(T) + d \log \Lambda + \frac{d}{2} \log \frac{\langle \Delta v(r) \rangle^*}{\pi T d} - d \\ &= S_c(T) + \frac{d}{2} \log \frac{\langle \Delta v(r) \rangle^*}{\pi T d} - \frac{d}{2}, \end{aligned} \quad (18)$$

where $S_c(T) = S_{\text{liq}}(T) + d \log \Lambda - d/2$ and $\langle \bullet \rangle^*$ denotes an average over the classical liquid at temperature $T^* = T/m$ (with $m = 1$ in the second line). These are exactly the results obtained by Mézard and Parisi in the classical case [28].

2.5 First order quantum correction

We now compute the first order quantum correction to the complexity in the semi-classical expansion. This can be done using a formal expansion in powers of \hbar [31]. For simplicity we will perform the computation directly for $m = 1$, starting from Eq. (15). The correction to the free energy of the liquid is

$$\delta F_{\text{liq}} = \frac{\Lambda^2}{24\pi} \langle \Delta v(r) \rangle^*, \quad (19)$$

then it is easy to see that

$$\delta [S_{\text{liq}}(T) - 2\beta K_{\text{liq}}(T)] = -\frac{\Lambda^2}{24\pi} \left[\frac{1}{T} \langle \Delta v(r) \rangle^* + \partial_T \langle \Delta v(r) \rangle^* \right]. \quad (20)$$

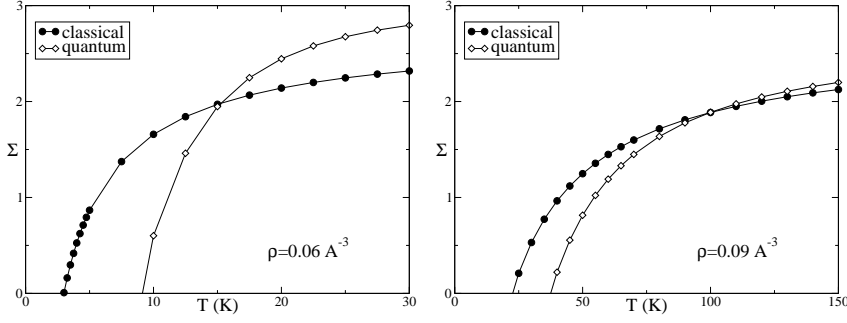


Fig. 1 The complexity $\Sigma_{\text{eq}}(T)$, both in the classical limit, Eq. (18), and with the quantum correction, Eq. (23), for two representative densities as a function of temperature.

To compute the variation of the free energy of the oscillator, we must take into account that the spring constant κ also has quantum corrections. From (19) we get $\delta g_0(r) = \frac{\Lambda^2}{24\pi} \Delta g_0(r)$, and using this we get

$$\frac{d}{2} \delta \kappa = \frac{\rho}{2} \int dr \delta g_0(r) \Delta v(r) = \frac{\rho}{2} \int dr \frac{\Lambda^2}{24\pi} \Delta g_0(r) \Delta v(r) = \frac{\Lambda^2}{24\pi} \langle \Delta^2 v(r) \rangle^* . \quad (21)$$

Denoting $\kappa_0 = 2 \langle \Delta v(r) \rangle^* / d$ and $\omega_0 = \sqrt{\kappa_0 / M}$ the classical spring constant and frequency, the first correction to the free energy of the oscillator is:

$$\delta[\beta d F_{\text{harm}}(\kappa)] = d \frac{(\beta \hbar \omega_0)^2}{24} + \frac{d}{2} \frac{\delta \kappa}{\kappa_0} = \frac{\Lambda^2}{24\pi T} \langle \Delta v(r) \rangle^* + \frac{d \Lambda^2}{48\pi} \frac{\langle \Delta^2 v(r) \rangle^*}{\langle \Delta v(r) \rangle^*} . \quad (22)$$

Collecting these results we obtain

$$\delta \Sigma(T) = \frac{\Lambda^2}{48\pi} \left[d \frac{\langle \Delta^2 v(r) \rangle^*}{\langle \Delta v(r) \rangle^*} - 2 \partial_T \langle \Delta v(r) \rangle^* \right] . \quad (23)$$

Note that using the results above one can check that at this order in \hbar we still have $2K_{\text{liq}} = d U_{\text{harm}}(\kappa)$ so the last term in (15) is still zero.

2.6 Results

We now present and discuss the results of the semiclassical expansion presented above. Note that although Eq. (23) has been derived in a formal expansion in powers of \hbar , we will in the following apply it using the real values of all physical constants. We focus on Helium 4, which we describe by means of a Lennard-Jones (LJ) potential $v(r) = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right]$ with $\epsilon = 10.22$ K and $\sigma = 2.556$ Å [32].

Given the Helium 4 mass, we have $\hbar^2 / (2M) = 6.0596$ K Å². The intensity of quantum fluctuations can be conveniently quantified by the ratio of the thermal wavelength and the inter-particle distance, $\Lambda^* = \Lambda \rho^{1/3} = \rho^{1/3} \sqrt{4\pi \hbar^2 / (2MT)} =$

$8.726 \text{ \AA K}^{1/2} \times \rho^{1/3} T^{-1/2}$. Although better potentials have been constructed [32], we choose the Lennard-Jones because it is the simplest and it gives reasonable quantitative results; in addition, the glass transition of a classical monoatomic Lennard-Jones system has been studied in great detail both analytically [28] and numerically [33], so we will be able to compare the results obtained in the classical case with the first order quantum correction.

To compute the classical equilibrium complexity and its quantum correction, according to Eqs. (18) and (23), we need the entropy of the classical liquid, as well as its $g(r)$, as a function of temperature. Note that since $m = 1$, the averages $\langle \bullet \rangle^*$ reduce to standard thermal averages over the classical liquid, that can be computed using $g(r)$. These quantities can be obtained using one of the integral equations of classical liquid theory [34]. Following [28], we choose the HNC closure [34] which has the advantage of providing a direct route to compute the free energy, and gives thermodynamic quantities with reasonable accuracy (of the order of 10%). The HNC equations for the LJ potential have been solved numerically using standard iteration schemes.

In Fig. 1 we report the complexity $\Sigma_{\text{eq}}(T)$ (also called configurational entropy in the context of structural glasses). The point where it vanishes marks the Kauzmann transition T_K to the ideal glass. We see that although the quantum correction to $\Sigma_{\text{eq}}(T)$ can be positive or negative, close to T_K it is always negative, therefore increasing the value of T_K with respect to the classical limit. At the lowest densities, T_K is found to increase by a factor of two with respect to the classical value. It is worth to note, however, that the semiclassical expansion is particularly bad for $\rho \leq 0.055 \text{ \AA}^{-3}$: in this region, the classical Lennard-Jones system enters the gas-liquid phase separation region [35], and the HNC equations cease to converge. Instead, for quantum Helium 4 the phase coexistence region is known to be much smaller, ending at $\rho \sim 0.022 \text{ \AA}^{-3}$ [36]. This is due to the large zero-point kinetic energy, and it signals that a semiclassical expansion is definitely not reasonable at low density, as expected.

In Fig. 2 (left panel) we report the glass transition temperature $T_K(\rho)$, obtained from the classical and the quantum computation. We see that at all densities, the quantum value is larger than the classical one (with the relative difference decreasing on increasing ρ). The classical value compares well with the value obtained from a numerical simulation of the classical LJ fluid [33]. The quantum value is below the melting temperature of Helium 4, as it should. Unfortunately, as already discussed, we cannot obtain results for the really interesting region of low density and temperature, where quantum effects are most important. For the classical LJ potential, T_K increases with density, therefore the transition line in the (ρ, Λ) plane is re-entrant. This is a trivial classical effect and has nothing to do with the re-entrance found in [22, 25, 23]. The important point, for the LJ case, is that quantum corrections increase the value of T_K , thereby promoting glass formation as stated in [22, 25, 23]. The region $\Lambda^* \sim 1$, where the semiclassical computation breaks down, is where we expect that the system should undergo a phase transition towards a superfluid state (the line $\Lambda^* = 1$ is reported in Fig. 2 for comparison). Since exchange was neglected from the very beginning, we cannot obviously analyze this transition.

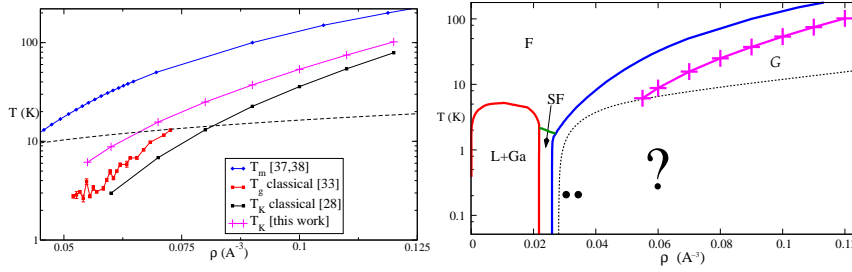


Fig. 2 (Color online) (*Left panel*) Glass transition lines $T_K(\rho)$, defined by $\Sigma_{\text{eq}}(T) = 0$, as obtained from the classical limit Eq. (18) [28] and including the first semiclassical quantum correction Eq. (23). Also shown is a numerical determination of the classical glass transition $T_g(\rho)$ from Ref. [33], and the numerically and experimentally determined melting temperature $T_m(\rho)$ of Helium 4, obtained from Refs. [36, 37]. The line $\Lambda^* = \Lambda\rho^{1/3} = 1$ below which quantum effects are very strong is plotted as a dashed line. (*Right panel*) Complete temperature-density phase diagram of Helium 4. *Equilibrium phases* (from Refs. [36, 37]): The red line delimits the liquid (L)-gas (Ga) coexistence region. The green line is the λ -line that separates the liquid from the superfluid (SF). The blue line is the melting transition below which the crystal phase is stable, and the disordered phase is metastable. *Glass phase*: The purple line is the glass (G) transition line obtained in the present work within the first order semiclassical expansion. The behavior of the amorphous phase, say, within the dashed black line is currently not well understood. The two black dots mark the state point investigated numerically in Ref. [6].

3 Speculations on the phase diagram of high density metastable Helium 4

In Fig. 2 (right panel) we report in the density-temperature plane¹ all the information now available for the phase diagram of Helium 4. We also draw the glass transition line that can be reached if crystallization can be avoided, see e.g. [38] for an experimental investigation of supercooled liquid Helium. Ref. [6] reports the existence of a metastable disordered superfluid state at $T = 0.2$ K and densities $\rho = 0.0292 \text{ \AA}^{-3}$ and $\rho = 0.0359 \text{ \AA}^{-3}$. These state points are marked by a black dot in Fig. 2. However, it is not clear to us whether this state is a liquid or a glass².

¹ The reason why we do not discuss the phase diagram in the more common pressure-temperature plane is that the computation of the pressure from the semiclassical expansion would introduce additional uncertainty on the position of the glass transition line. Moreover, both theoretical and numerical computations are more naturally done at constant density.

² In Ref. [6] the system was named a glass because it was observed that a non-homogeneous density profile lasted for a certain time during a Path Integral Monte Carlo (PIMC) simulation. However, although certainly very stimulating, we find that these results are not conclusive because of the following reasons:

- PIMC simulations only give access to a fictitious classical dynamics of a system of polymers that represent the imaginary time evolution of the quantum particles. Despite some claims that this dynamics might be close to the true Schrödinger dynamics of quantum particles (see e.g. [22]), the connection between the two has never been convincingly shown.
- We don't find the evidence reported in Ref. [6] strong enough to support the existence of a glass phase, even for the PIMC polymers. Our point is that, by analogy with simpler classical systems, in order to establish the presence of a glass phase one has to perform a careful systematic study of time-dependent density correlations, and show that such correlations develop a plateau at intermediate time scales, that there is a clear separation of time scales between a short time (so-called β) and long time (so-called α) relaxation, and that there is aging in the glass phase. In absence of these evidences, avoided or interrupted nucleation of

As already stressed, our results for the glass (purple) transition line are reliable only in the high density and high temperature regime in the (ρ, T) phase diagram, where a semiclassical expansion makes sense. Since our theory is unable to include exchange correctly, we cannot describe the interplay between superfluidity and glassiness taking place at low density and low temperature, say, within the dashed black line in the phase diagram of Fig. 2. In the rest of this section we describe two possible scenarii for the transitions taking place in the region corresponding to the question mark.

3.1 A first order superfluid transition: the heterogeneous superglass phase

In Fig. 3 (left panel) we report a conjectural phase diagram for metastable Helium 4 in the (ρ, T) plane, as it emerges by the combination of Refs. [22, 23, 24] and the present study. The purple line is the glass transition line obtained in the present work within the first order semiclassical expansion. Below it, the system is a glass. In [23] it was predicted that the λ -line might become a first order transition line at high density. Although this prediction has been made for a quite abstract lattice mean field model of dense liquids, it is interesting to discuss it since these models in several cases display a phenomenology that is very similar to particle systems. One should then observe a superfluid (SF)-glass (G) coexistence, leading to an heterogeneous superglass where regions of low-density superfluid would coexist with regions of high-density glass. The reader should keep in mind that the qualitative behavior (in particular the form) of the λ -line in the metastable phase is entirely conjectural. Moreover we currently don't have any estimate of the exact location of this line; therefore, its position in Fig. 3 is arbitrary.

3.2 The homogeneous superglass phase

A second scenario is that the λ -line always continues to be a second order transition in the metastable part of the phase diagram. In this case, it will very probably cross the glass transition line at some point, leading to a *superglass* phase (SG). This picture, which is displayed in Fig. 3 (right panel), is consistent with two quite different physical scenarios for the glass transition line at low temperature:

- In Refs. [39, 40], a phase diagram like the one in Fig. 3 (right panel) was found. However, in the models investigated there, the glass transition is continuous, like for spin glasses: the amplitude of frozen-in density fluctuations changes continuously at the transition. This is very different from the transition of structural glasses, that we actually find in our semiclassical analysis, and that corresponds to a discontinuous jump of the amplitude of the frozen-in density fluctuations, the transition remaining however of second order. To

a polycrystalline phase can easily be confused with the formation of a true glass, as it has been shown in a number of studies on classical systems. An example of this in the context of distinguishable Helium 4 particles was given in Ref. [15].

For this reason, the system of Ref. [6] might be in reality a moderately viscous supercooled liquid, and not a really arrested glass. We hope that further numerical work will clarify this issue.

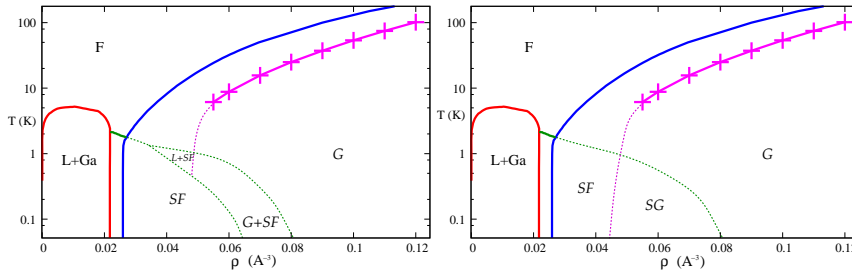


Fig. 3 (Color online) (*Left panel*) A conjectural phase diagram for metastable Helium 4 as it emerges from the combination of Refs. [22, 23, 24] and the present study. The stable part of the phase diagram is as in Fig. 2. The metastable phases are indicated in *italic*. The purple line is the glass transition line obtained in the present work within the first order semiclassical expansion. The λ -line becomes, at some point inside the supercooled liquid, a first order transition line [23]. One should therefore observe a superfluid (SF)-glass (G) coexistence. Note that we currently do not have any quantitative estimate of the behavior of the λ -line inside the metastable phase, which is drawn at arbitrary position in the figure. (*Right panel*) Another possible phase diagram for metastable Helium 4. Same colors and remarks as in the caption of Fig. 3. The λ -line always stays a second order transition. When it crosses the glass transition line, a superglass (SG) phase appears.

match these two findings one can conjecture that below some temperature the glass transition line ceases to be discontinuous and becomes continuous instead. If this is the case, at low temperature glassy amorphous order would grow gradually from the liquid, and the relaxational dynamics approaching the glass transition would be very different from the one of classical supercooled liquids.

- The second possibility is that the glass transition line continues to be discontinuous down to $T = 0$. This would imply the existence of a quantum discontinuous glass transition as a function of density. This has indeed been found in [21], based on a variational Jastrow description of the ground state of an interacting quantum particle system. Still some conceptual problems remain: indeed, all mean field models that could be solved exactly and that display such a transition in the classical limit, show that the latter becomes first order at $T = 0$ [16, 17, 18, 19, 20]. The model of [21] displays such a transition, but its ground state wavefunction has only short-ranged two-body correlations, and therefore can be mapped onto a classical liquid; this is likely not to be the case for more generic models. A better understanding of perturbation theory around Jastrow wavefunctions, which was only attempted in [21], would be very helpful.

4 Discussion

Before concluding the paper, we critically examine the assumptions on which our method was based: we highlight its weaknesses and we discuss how to improve it.

The first important issue that has to be discussed is the coupling between replicas. In section 2, Eq. (2) we have coupled the replicas as in Ref. [28], which is appropriate in the classical case. Here we discuss whether this way of cou-

pling replicas remains correct in the quantum case. In order to discuss this issue we go back to the original derivation of Monasson [26]. Since the glass is characterized by an inhomogeneous density profile, Monasson considered a system whose density is coupled to a static reference density field $\sigma(r)$. We denote by $\rho(r; \{x(\tau)\}) = \sum_i f(r - x_i(\tau))$ a smoothed density profile at imaginary time τ , where the function $f(r)$ is normalized to 1 and very short ranged. Then in the path integral representation we get

$$Z[\sigma] = \int \mathcal{D}\{x\} e^{-S_0[\{x\}] - \varepsilon \int dr d\tau [\sigma(r) - \rho(r; \{x(\tau)\})]^2}, \quad (24)$$

where $S_0[\{x\}]$ is the unperturbed action and the trajectories x_i are assumed to be periodic in $\tau \rightarrow \tau + \beta$; a summation over the permutations due to Bose statistics is implicit. Following [26], we consider the partition function of the field σ with ‘‘Hamiltonian’’ $F[\sigma] = -T \log Z[\sigma]$ at inverse temperature βm , given by $Z_m = \int \mathcal{D}\sigma(r) \exp[-\beta m F[\sigma]] = \int \mathcal{D}\sigma(r) Z[\sigma]^m$. Integrating over $\sigma(r)$ we get:

$$Z_m = \int \mathcal{D}\{x^a\} e^{-\sum_a S_0[\{x^a\}] - \frac{\varepsilon}{2\beta m} \int d\tau d\tau' dr \sum_{ab} [\rho(r; \{x^a(\tau)\}) - \rho(r; \{x^b(\tau')\})]^2}. \quad (25)$$

Defining $F(r - r') \equiv \int dz f(r - z) f(z - r')$ (note that $F(r)$ is still a normalized short-range function), the coupling term has the form

$$\begin{aligned} d_{ab}(\tau, \tau') &= \int dr [\rho(r; \{x^a(\tau)\}) - \rho(r; \{x^b(\tau')\})]^2 \\ &= \sum_{ij} [F(x_i^a(\tau) - x_j^a(\tau)) + F(x_i^b(\tau') - x_j^b(\tau')) - 2F(x_i^a(\tau) - x_j^b(\tau'))], \end{aligned} \quad (26)$$

This coupling makes the configuration $x_i^a(\tau)$ similar to $x_j^b(\tau')$ or to a permutation of it. Thus, in the strong coupling limit, the particles build ‘‘molecules’’ made of one atom of each replica. *If we make the approximation of ignoring permutations of the atoms of one replica belonging to different molecules*, or in other word if we only allow permutation of entire molecules (molecules are Bosons but individual atoms cannot be exchanged), then we can re-label the atoms in such a way that molecule 1 is built by x_i^a , and so on. In (26), due to the short range of $F(r)$ we can neglect all terms that couple atoms with $i \neq j$ and we get

$$d_{ab}(\tau, \tau') \sim 2 \sum_i [F(0) - F(x_i^a(\tau) - x_i^b(\tau'))] \sim - \sum_i F''(0) (x_i^a(\tau) - x_i^b(\tau'))^2,$$

where we used that $F(r)$ is short-ranged and the coupling is large, so that atoms in a molecule will vibrate around the bottom of the potential well defined by $F(r)$, and we can assume that $x_i^a(\tau) - x_i^b(\tau')$ is small; the result is that the coupling in a molecule can be assumed to be harmonic (note that $F''(0) < 0$). Inserting this result in the path integral (25), we almost recover the path integral of the molecular system described by the Hamiltonian (2), except for the fact that replicas are coupled at different times. We therefore have to make the further approximation of *ignoring the coupling of replicas at different imaginary times*.

The above derivation of our starting point, Eq. (2), is then based on two crucial approximations:

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- *We neglected exchange of atoms between different molecules.* This is incorrect when exchange is important. But taking into account these processes, the molecules lose their identity. A coupling term such as the one in (2) cannot be written since it explicitly breaks permutation symmetry of each replica. Therefore one should write a coupling that is manifestly invariant under permutation of the atoms of each replica, independently of the other replicas. This can be done, but solving the Hamiltonian looks a much more difficult task.
 - *We neglected the non-local coupling of replicas in imaginary time.* It is possible that since the limit $\varepsilon \rightarrow 0$ must be taken in the end, the precise form of the replica coupling is not relevant. It would be nice to perform the calculation including the non-local coupling, but this is technically more involved since a Hamiltonian formulation is not possible in this case. One has to work directly with path integrals. We attempted to perform such a computation but did not succeed.

We note that although these problems seem very serious, they should not matter in the semiclassical regime where exchange can be neglected, and quantum fluctuations are small, so that the path integral is over a short time and coupling the replicas at the same time or at different times should not change the result a lot. Therefore, we hope that Eq. (2) is justified as the starting point of our semiclassical calculation.

Another major source of concern is that a kinetic energy contribution to the equilibrium complexity appears in Eq. (15). This contribution obviously vanishes in the classical case, and luckily enough it still vanishes at first order in the semiclassical expansion. However, it is definitely non-zero beyond this order. This is problematic since it would imply that the kinetic energy of the liquid and the glassy states visited at $m = 1$ are not the same, which is instead a basic premise of the replica approach to glasses (all correlation functions should actually be the same). The issue is not the replica approach since the original Monasson's derivation can be formally generalized to the quantum case by using the density field $\rho(r; \{x(\tau)\})$ without encountering any difficulty, as we discussed above. The problem instead relies in the hypothesis behind the cage expansion, in particular the harmonic approximation. It is not clear to us how to find a solution. Possibly, a more refined description of the liquid and a more accurate expansion in ε should at least reduce the problem quantitatively.

5 Conclusions

We presented a first attempt to build a replica theory of quantum glasses generalizing the recipe of [28] to the quantum case, and performing a semiclassical expansion that seems to give meaningful results at least for high density. In particular, we estimated the glass transition line and found that, as already reported in [22, 23, 24, 25], quantum fluctuations promote glass formation. We critically discussed some issues related to our approach, in particular the role of exchange and a technical problem related to the kinetic energy. Although a full theory of quantum glasses must include these effects to provide meaningful results, they are not expected to be important in the semiclassical regime we focused on.

Based on the combination of the present study and previous ones [22, 23], we

conjectured a phase diagram of dense metastable Helium 4 which is reported in Fig. 3 (left panel). In this case, it might well be that the glass ceases to exist in the strongly quantum region, since it undergoes a first order transition to the superfluid; then, the present treatment could be appropriate to describe the glass, and the problem would be then to estimate the free energy of the dense metastable liquid in order to look for a first order transition. We discussed in Fig. 3 (right panel) another possible scenario, in which the glass phase becomes a superglass and a zero-temperature glass transition exist. The nature of this quantum glass transition remains unclear.

There are several ways to go beyond our treatment.

- One could try to use influence functional methods, which were used in [22] to compute the thermodynamical properties of the liquid. Although these methods do not include exchange, they should allow for a more accurate description of the glass.
- Another possibility would be to use finite-temperature variational methods [41] in order to extend the computation of [21] to finite temperature. This allows one to include exchange, but not to describe the normal phase correctly.
- A careful investigation of the perturbation theory around the special model of [21] should help to understand if a quantum discontinuous glass transition indeed takes place.

However, since the physical picture is very far from being settled, before pushing forward the theory it would be extremely interesting to perform Quantum Monte Carlo simulations or experiments (along the lines of [38]) in the dense metastable liquid region, to investigate the behavior of the λ -line at high density.

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