Critical temperature of the superfluid transition in Bose liquids

S. M. Apenko*

I.E. Tamm Theory Division, P.N. Lebedev Physical Institute, Moscow 117924, Russia

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A phenomenological criterion for the superfluid transition is proposed, which is similar to the Lindemann criterion for the crystal melting. Then we derive a formula for the critical temperature, relating T_λ to the mean kinetic energy per particle above the transition. The suppression of the critical temperature in a sufficiently dense liquid is described as a result of the quantum decoherence phenomenon. The theory can account for the observed dependence of T_λ on density in liquid helium and results in an estimate $T_\lambda \sim 1.1$ K for molecular hydrogen. [S0163-1829(99)08529-X]

In connection with the search for new bose-condense systems, attention was again attracted recently to the possible superfluidity in molecular hydrogen H_2,[1,2] discussed earlier in Refs. 3–5. The very possibility crucially depends on the expected value of the critical temperature T_λ, since normally molecular hydrogen crystallizes at ~ 14 K and some measures should be taken to keep it in liquid phase at lower temperatures, which may appear harder to achieve if T_λ is too low. Hence one needs first to evaluate T_λ in supercooled hydrogen. In a more general context we are faced with the problem of determining T_λ in a given quantum liquid. Usually, the first step is to use the well-known formula for the Bose-Einstein condensation temperature in the ideal gas,

$$T_\lambda \simeq 3.31 \frac{\hbar}{m} n^{2/3}, \qquad (1)$$

where n is the density and m is the particle mass ($\kappa_B = 1$). Though this formula results, e.g., in a rather reasonable estimate of T_λ in liquid helium (~ 3 K instead of the correct value 2.17 K at the saturated vapor pressure) it still seems unsatisfactory in case of dense liquids because it cannot account for the observable dependence of T_λ on density. According to Eq. (1) the critical temperature increases as $n^{2/3}$, while, on the contrary, T_λ in liquid ^{4}He slightly decreases when the system is compressed. Certainly, one can argue that it is not the bare mass that enters in Eq. (1) but rather an effective one, depending on density,[6] but there seems to be no simple and general expression for this effective mass. A calculation of T_λ based on the Landau quasiparticle spectrum[7] can be applied only when this spectrum is already known. Qualitatively the observed behavior of T_λ can be explained also in the lattice model[8] but in general it is not so obvious how to relate the properties of a liquid to those of bosons on a lattice. For this reason some other estimate of T_λ is required, simple enough to serve as a first approximation, but which can account for nonmonotonic dependence of the critical temperature of a bose system on its density. In this article we propose such a general formula, which relates T_λ to the mean kinetic energy per particle in the normal phase— the quantity which behavior is now well understood.[9–12]

The most general approach to superfluidity is, perhaps, to start with the Feynman path-integral expression for the partition function Z for a system of N interacting bose particles:[6]

$$Z = \frac{1}{N!} \sum_P \int \prod_i d\mathbf{r}_i \int \prod_i \mathcal{D}\mathbf{r}_i(\tau) \exp\left(-\frac{1}{\hbar}S\right), \qquad (2)$$

$$S = \int_0^{\hbar\beta} \left[\frac{m\dot{\mathbf{r}}_i^2}{2} + \sum_{i<j} V(\mathbf{r}_j - \mathbf{r}_j) \right] d\tau,$$

where $V(\mathbf{r}_i - \mathbf{r}_j)$ is the interparticle interaction potential and $\beta = 1/T$. The integration in Eq. (2) is over all paths with $\mathbf{r}_i(0) = \mathbf{r}_i$, $\mathbf{r}_i(\hbar\beta) = P\mathbf{r}_i$, where P is some permutation of N particles, and the sum in Eq. (2) is over all such permutations.

At high temperatures only the identity permutation is important, since particles cannot move far away from their initial positions in ''time'' $\hbar\beta$. As β increases, a given path $\mathbf{r}(\tau)$ can spread on a larger distance, until suddenly it appears possible to end the path at the position of a neighboring particle. Then, as discussed in detail in the recent review,[13] the rings of exchanges of arbitrary length are formed, which can be shown to lead to the superfluid behavior. Let us now consider a system of *distinguishable* particles, take arbitrary particle and evaluate its mean-square displacement from initial position $\langle (\delta\mathbf{r})^2 \rangle$ in imaginary time. To estimate the critical temperature, when exchanges can no longer be neglected, we propose a criterion

$$\langle (\delta\mathbf{r})^2 \rangle \equiv \frac{1}{\hbar\beta} \int_0^{\hbar\beta} \langle [\mathbf{r}(\tau) - \mathbf{r}(0)]^2 \rangle d\tau = \xi a^2, \qquad (3)$$

where ξ is some numerical factor, to be determined later. This condition merely states that the mean displacement of a given particle in imaginary time is comparable to the interparticle spacing. The condition (3) is similar in spirit to the well known Lindemann criterion in case of crystal melting and is inspired by the visual representation of the paths, arising from numerical simulations.[13]

In the ideal gas the left hand side of Eq. (3) is essentially the square of the de Broglie thermal wavelength $\lambda_T^2 = \hbar^2/mT$, but interactions will tend to reduce $\langle (\delta\mathbf{r})^2 \rangle$ (this was observed, e.g., in Ref. 14). There are two main mechanisms of such a reduction. The first one is related to decoherence due to interaction with environment. Neighboring particles, in a sense, ''measure'' the position of the particle we are looking at, thus reducing its quantum uncertainty in

coordinate space. Such a decoherence phenomenon was discussed a lot for more than two decades with respect to the transition from the quantum behavior to the classical one (see, e.g., Ref. 15). The second mechanism is more typical for crystals or glasses, where particles are almost localized by potential barriers.

The problem of estimating of $\langle(\delta\mathbf{r})^2\rangle$ in a system of interacting particles is still a very complicated one, even if exchanges are neglected. It can be significantly simplified, however, by treating the rest of the system as some simple fluctuating environment. The simplest choice is the Caldeira-Leggett model,[16] which describes interaction of a particle with a thermal bath of harmonic oscillators. In this model the particle motion in the imaginary time is governed by the effective action

$$S = \int_0^{\hbar\beta} d\tau \frac{m\dot{\mathbf{r}}^2}{2} - \frac{1}{4}\int_0^{\hbar\beta} d\tau$$

$$\times \int_0^{\hbar\beta} d\sigma K(\tau-\sigma)(\mathbf{r}(\tau)-\mathbf{r}(\sigma))^2, \qquad (4)$$

where the kernel $K(\tau)$ is determined by

$$K(\tau) = (m/\hbar\beta)\sum_{n=-\infty}^{+\infty} \zeta_n e^{i\omega_n\tau},$$

$$\zeta_n = \frac{1}{m}\int_0^\infty \frac{d\omega}{\pi}\frac{I(\omega)}{\omega}\frac{2\omega_n}{\omega^2+\omega_n^2}, \qquad (5)$$

where $\omega_n = 2\pi n/\hbar\beta$ and $I(\omega)$ is the spectral density of bath oscillators (see, e.g., Ref. 17). In our case one can view Eq. (4) as a trial action and try to evaluate $K(\tau)$ variationally, but it is easier to directly relate the parameters of the effective action to some observables of the system (see below). For quadratic action (4) in three dimensions the mean displacement is

$$\langle(\delta\mathbf{r})^2\rangle = \frac{12}{m\beta}\sum_{n=1}^\infty \frac{1}{\omega_n^2+\zeta_n}. \qquad (6)$$

At small frequencies it seems natural to expect the Ohmic behavior of the kernel $K(\tau)$, corresponding to the linear friction, when damping is proportional to the velocity of the particle. This is modeled by $I(\omega) = \gamma m\omega$, where γ is the effective damping parameter and $\zeta_n = \gamma|\omega_n|$. The Caldeira-Leggett model with such a dissipation kernel is a quantum analog of the standard Langevin equation with the friction $m\gamma$ and with the white-noise random force. Thus we treat the particle motion as that of a Brownian particle to model the decoherence effect. We expect this approach to be qualitatively adequate when $\langle\delta\mathbf{r}^2\rangle$ is less than the square of the interparticle spacing. Then the sum in Eq. (6) is carried out and

$$\langle(\delta\mathbf{r})^2\rangle = \frac{6}{\pi}\frac{\hbar}{m\gamma}\left[C+\psi\left(1+\frac{\hbar\gamma\beta}{2\pi}\right)\right], \qquad (7)$$

where $C\simeq 0.577\ldots$ is Euler's constant and $\psi(x)$ is the psi function (the logarithmic derivative of the gamma function).

Now we have only one parameter, γ, which describes interactions in the system. The ideal-gas limit is recovered at $\gamma\to 0$, when $\langle(\delta\mathbf{r})^2\rangle\to\hbar^2\beta/2m$. Then the criterion (3) results in $T_\lambda = (1/2\xi)\hbar^2 n^{2/3}/m$. Since this is the ideal-gas formula for the critical temperature (1) we conclude that $1/2\xi\simeq 3.31$, i.e.,

$$\xi\simeq 0.15. \qquad (8)$$

Though the criterion (3) is useful only if ξ is some universal constant we cannot exclude some weak dependence of ξ on density. We expect that ξ may be slightly smaller than Eq. (8) in a system with short-range order since it is more likely for a particle in a liquid to find a neighbor for exchange at a suitable distance than in the ideal gas, where density fluctuations are more important.[20]

In the opposite case, when $\hbar\gamma\beta/2\pi\gg 1$, the mean displacement diverges logarithmically, i.e., $\langle(\delta\mathbf{r})^2\rangle\sim\ln(\hbar\gamma\beta/2\pi)$. Hence $\langle(\delta\mathbf{r})^2\rangle$ is much less than the de Broglie thermal wavelength of a free particle. This logarithmic behavior is related to the Ohmic spectrum at small frequencies (cf. Refs. 18 and 19). Then for the critical temperature at $\gamma\to\infty$ we have a very simple formula:

$$T_\lambda = \alpha\frac{\hbar\gamma}{2\pi}\exp\left(-\xi\frac{\pi}{6}\frac{\hbar\gamma}{T_0}\right), \quad T_0 = \frac{\hbar^2}{m}n^{2/3}, \qquad (9)$$

where $\alpha = \exp(C)$.

We see now, that the temperature of the superfluid transition crucially depends on the ratio $\hbar\gamma/T_0$. At $\hbar\gamma\ll T_0$ the estimate (1) is valid and the critical temperature is essentially $T_0\sim n^{2/3}$, while at $\hbar\gamma\gg T_0$ the critical temperature of the λ transition is exponentially small due to the decoherence phenomenon. Qualitatively this can be understood as follows: in the process of exchange particles move through the viscous media and lose the coherence needed for superfluidity to establish. Since γ should increase with increasing density, the formula obtained do describe the suppression of T_λ in sufficiently dense systems.

The approximation of a constant friction would be correct if we dealt with a heavy Brownian particle, which moves more slowly than particles in a liquid. In real liquids the Ohmic spectrum $I(\omega)\sim\omega$ is physical only at frequencies lower than the collision rate and must have a cutoff at some frequency ω_c, which now should be taken into account. One can take, e.g., the Drude model for the damping:[17]

$$\zeta_n = \gamma\omega_c|\omega_n|/(\omega_c+|\omega_n|). \qquad (10)$$

Apart from the dispersion of the friction coefficient (memory effects) this implies that the random force acting on a particle is correlated for times less than $1/\omega_c$.

Qualitatively, however, the picture outlined above remains unchanged. If we, e.g., put $\omega_c = \gamma$ then again the formula (9) is recovered but now with $\alpha = \exp(C+\sqrt{3}\pi/9)$.

At low temperatures γ can be related to the mean kinetic energy per particle. The kinetic energy in the Caldeira-Leggett model may be written as

$$K(T) = \frac{3}{2}T + \frac{3}{\beta}\sum_{n=1}^\infty \frac{\zeta_n}{\omega_n^2+\zeta_n}. \qquad (11)$$

The sum is easily evaluated for a model (10) and, e.g., at $\omega_c\to\infty$ (purely Ohmic limit) we obtain

$$K(T) = \frac{3}{2}T + \frac{3\hbar\gamma}{2\pi}\left[\ln\frac{\hbar\omega_c\beta}{2\pi} - \psi\left(1 + \frac{\hbar\gamma\beta}{2\pi}\right)\right]. \quad (12)$$

This very expression was derived earlier[10] for liquid ^{4}He above the λ point by taking the velocity autocorrelation function to be of a simple exponential form with damping γ (denoted in Ref. 10 by ω_0), just as for the Brownian particle. At $T \gg \hbar\gamma$ the kinetic energy tends to $\frac{3}{2}T$, while at low temperatures $K(T)$ has a finite limit, denoted hereafter by $\dot{K}$. This is quite a general behavior, valid in the Debye model as well.[22] Strictly speaking, K is the kinetic energy just above T_λ, since exchanges are not taken into account, but, e.g., in helium the difference between $K \sim 16$ K and the real zero-point energy is ~ 1.5 K and will be neglected here. For the particular model (12) we have $K = (3\hbar\gamma/2\pi)\ln(\omega_c/\gamma)$, i.e., as was already mentioned in Ref. 10 (see also Ref. 21), $\hbar\gamma$ up to a logarithmic factor coincides with K. Thus we conclude that in general case at $T \ll K$ (but above the transition)

$$\hbar\gamma \sim K. \quad (13)$$

For different $I(\omega)$ the proportionality coefficient is actually cutoff dependent and, e.g., for the model (10) with $\omega_c = \gamma$ one has $\hbar\gamma = \sqrt{3}K$. Since in the formula (9) the factor α also depends on a high-frequency behavior of $I(\omega)$ we can finally write

$$T_\lambda = A\,K\exp\left(-B\frac{K}{T_0}\right) \quad (14)$$

for $K \gg T_0$, where A and B are some model dependent constants. For the Caldeira-Leggett model with damping kernel (10) at $\omega_c = \gamma$ we have

$$A = (\sqrt{3}/2\pi)\exp(C + \sqrt{3}\pi/9) \simeq 0.899,$$

$$B = (\sqrt{3}\pi/6)\,\xi \simeq 0.907\,\xi. \quad (15)$$

The physical meaning of Eq. (14) is clear: if K, which at $T \ll K$ may be viewed as an effective "internal" temperature of the system,[10] is much larger than the transition temperature in the ideal gas, the superfluidity is suppressed.

Let us now compare the formula (14) with experimental data for liquid helium. We need then some explicit expression for $K(n)$. There are different estimates of zero-point energy in helium (see, e.g., Refs. 23 and 10). All of them are in general consistent both with the experimental data[11] and with results of path integral Monte Carlo (PIMC) calculations.[9,12] The kinetic energy increases with density due to the repulsion core in interatomic potential. Here we shall use the London's formula

$$K = \frac{2\pi\hbar^2 d}{m(a - 0.891d)^2(a + 0.713d)}, \quad (16)$$

where $a = n^{-1/3}$, $d \simeq 2.4$ Å.[23] This formula is simple and transparent being an interpolation between low-density limit $K \sim \hbar^2 dn/m$ of the energy of a gas of hard spheres of radius d, and a quantum-mechanical estimate $K \sim \hbar^2/m(a - d_0)^2$ with $d_0 \sim d$ in the high-density limit.

With this expression for K the formula (14) with coefficients (15) at $\xi = 0.15$ results in $T_\lambda \simeq 1.35$ K, which is lower

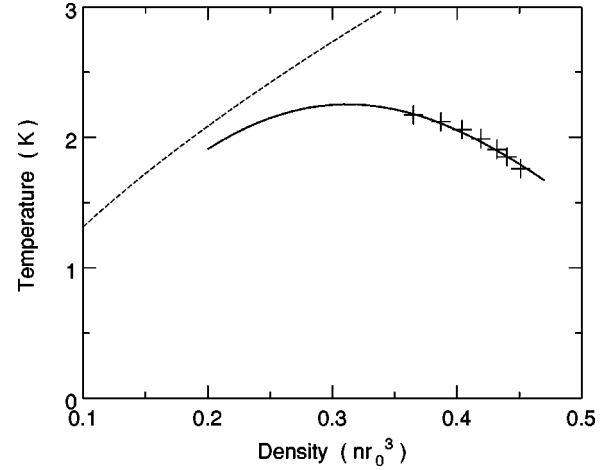FIG. 1. Temperature of the superfluid transition vs the reduced density $n^* = nr_0^3$ ($r_0 = 2.556$ Å). Solid line is the theory at $\xi = 0.12$, crosses denotes experimental data for helium, dashed line corresponds to the ideal gas.

than the experimental value of 2.17 K. We may recall, however, that ξ may be smaller in a dense system than in the ideal gas, and try to fit Eq. (14) to experimental data treating ξ as an adjustible parameter. The result of such a fit is shown in Fig. 1. Here the critical temperature is shown as a function of the reduced density $n^* = nr_0^3$, where $r_0 = 2.556$ Å is the length parameter of the Lennard-Jones interatomic potential for helium. Experimental values of T_λ for liquid helium, taken from the very accurate empirical expression for the λ line,[24] are shown by crosses. The dashed curve represents the ideal-gas formula (1), while the solid one is the best fit of T_λ from Eqs. (14) and (15) to experiment, which corresponds to $\xi \simeq 0.12$. This value is only slightly smaller than the ideal-gas limit $\xi = 0.15$. Given the simplicity of the assumptions, the agreement of the theory with experiment seems quite satisfactory. Since $BK/T_0 \sim 1$ at $n^* \sim 0.2$, we cannot expect Eq. (14) to be quantitatively valid at such a low density, but still it is clear that the theory really can describe the crossover from the ideal-gas behavior to the observed dependence of T_λ on the density.

Now, as far as the molecular hydrogen is concerned, we may take the London's formula (16) with $d \simeq 2.7$ Å as estimated in Ref. 25, which corresponds to a stronger interparticle potential. Assuming the numerical coefficients A and B to be the same as in helium (with $\xi \simeq 0.12$) we obtain from Eq. (14) $T_\lambda \simeq 1.1$ K for density $n \simeq 26$ nm^{-3}. Even if we reduced the density to that of helium, i.e., $n \simeq 22$ nm^{-3} the critical temperature would be only ~ 2.1 K. This is much less optimistic than the original estimate 6–8 K,[3] based on the ideal-gas formula (1), though consistent with later estimates[5] and with the PIMC analysis of finite hydrogen clusters.[26]

In conclusion, we have obtained a general estimate for the critical temperature of the superfluid transition in a bose liquid. Starting from the phenomenological Lindemann-like criterion (3) for the transition and modeling the decoherence effect, which suppresses T_λ in liquid, by the Caldeira-Leggett model, we arrive at a simple expression (14), relating T_λ to the kinetic energy K (which is essentially the zero-point energy per particle). The fast increase of K with density due to the repulsion core in interatomic potential accounts

then for the reduction of T_λ in dense systems, when $K \gg T_0$ $\sim \hbar^2 n^{2/3}/m$, as observed, e.g., in liquid helium. Using the London's interpolation formula for $K(n)$ and one fitting parameter the model can even quantitatively describe experimental behavior of T_λ.

Our approach suggests that, in a liquid, contrary to weakly nonideal gas, T_0 may not correspond to any characteristic temperature. The temperature where quantum effects become important is of the order of K, and is much larger than T_0,[10] while T_λ at high densities is smaller than T_0, and the difference increases with density. The error of using the ideal-gas formula (1) is not so much for helium, but may be of importance in molecular hydrogen (which is more dense, due to stronger interaction), where our formula gives much smaller value of T_λ.

An important question, which remains open within the phenomenological approach, concerns the universality of the numerical constants in Eq. (14). Though the parameter ξ (analogous to the Lindemann ratio in case of crystal melting) is to some extent fixed by the ideal-gas limit (8) further work is needed to clarify its possible dependence on system parameters.

*Electronic address: apenko@td.lpi.ac.ru

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