The connection between collective diffusion in classic liquids and the superfluidity phenomenon in quantum liquids

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This study suggests the mechanism for atomic motion in quantum liquids at low temperatures where the lifetime of the states that are responsible for atomic motion becomes macroscopically long. This mechanism is an analog to the motion of crowdions in the one-dimensional Frenkel-Kontorova model. Soliton-like atomic motion along linear directions, and the circular motions provide a means of explaining many macroscopic phenomena occurring at the transition of ^{4}He to a superfluid state, such as the behavior of rotating superfluid helium and the flow potentiality of the superfluid phase. The thermodynamics of rotating ^{4}He is considered under the assumption that Bose-Einstein condensate is not the ground state of Bose-Einstein liquid. An experiment is suggested in order to validate our approach.

Keywords: superfluid ^{4}He, phase transition, string-like diffusion, thermodynamics of rotating ^{4}He, Bose-Einstein condensation, Frenkel-Kontorova model, supersolid phenomenon.

1. Introduction

A great body of research has been done to date, lending support to the view that in classic liquids, including at low temperatures, an elementary act of atomic motion has a collective character (the term “string-like diffusion is commonly accepted in English-language publications) [1-8]. The results reported in the just cited references were obtained via numerical computer experiments which are rather difficult to perform in the region of low temperatures where quantum effects manifest themselves.

Theoretical research into quantum liquids [9–11] done from first principles, where the gas of noninteracting particles is taken as a zeroth-order approximation, cannot be recognized fundamentally as mathematically rigorous, because the parameters of density and interaction force were far from their values in real liquids. Therefore, the advancement of condensed-matter physics benefited greatly from heuristic approaches; for instance, as early as 70 years ago L.D.Landau [10] postulated a two-fluid model to obtain phenomenological equations which, subsequently, were never excelled in universality of the description of superfluid ^{4}He. Furthermore, the phenomenological Ginzburg-Landau equation describing superconducting

fluids in metals is used in practice much more frequently than all the other equations which are obtained from first principles.

It is our belief that the main difficulties encountered in describing liquids are due to the improper choice of a zeroth-order approximation in the form of the gas of noninteracting particles. This approximation describes reasonably well the equilibrium thermodynamic parameters of a fluid in which the phonon modes are of first importance. But the processes of atomic motion are very inconvenient to describe in terms of phonons. One is led to suggest that to describe a condensed state required that the main element of a given phenomenon be guessed right, i.e. the ground state from which the energy of elementary excitations is reckoned. We suggest an explanation for the mechanism of mass motion in quantum liquids in which the ground state of Bose-Einstein liquid is represented not by Bose-Einstein condensate with zero momentum but by a chaotic network of linear chains of a macroscopic scale along which crowdions travel without friction. Next, we consider the effects arising in the event of discarding the assumption that the superfluid phase is identical with Bose-Einstein condensate.

2.The hypothesis about the physics of the superfluid phase

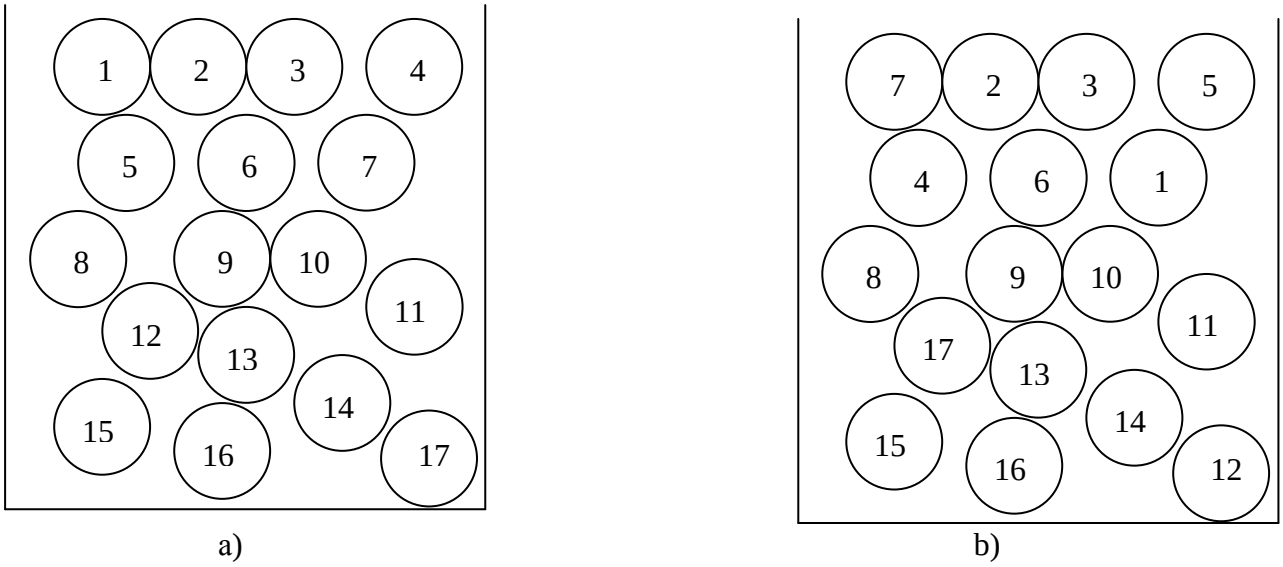
The objective of this paper is to describe qualitatively the microscopic mechanism of superfluidity in Bose-Einstein liquids, drawing on currently available theoretical and experimental data. The motivation for this study is that fundamental studies of superfluidity examined the possible energy spectrum of excited states of superfluid liquid rather than the physics of the ground state. This may well lead to paradoxical conclusions. In, for example, [10, p 115] we can find L. Landau's reasoning: “An important property of the superfluid flow is its potentiality:

$$rotV_s = 0\,, \qquad (1)$$

where V_s is the superfluid phase velocity. This property is the macroscopic expression of the fact that elementary excitations with a long wavelength (i.e. with small momenta) are phonons”. But phonon (oscillatory) and roton (rotational) motions cannot transport the mass and, hence, atomic motions cannot be described within the frames of excitations of these types! The assumption that the superfluid phase is Bose-Einstein condensate (a coherent quantum state with zero momentum) does not permit the hypermobility of atoms in the ground state to be explained.

What is the superfluid phase (SP)? If we cool ^{4}He to a temperature, however low where the number of excitations of the ground state is negligibly small, then, just the same, it does not lose its superfluidity; that is to say that atomic motion is not generated by excitations. As was demonstrated by R. Feinman [9], describing atomic motion in a quantum liquid is not an easy matter. In fact, the two configurations for distinguishable particles in Fig. 1 are obtained one from another by displacing atoms; in the case of Bose-Einstein statistics, these configurations are

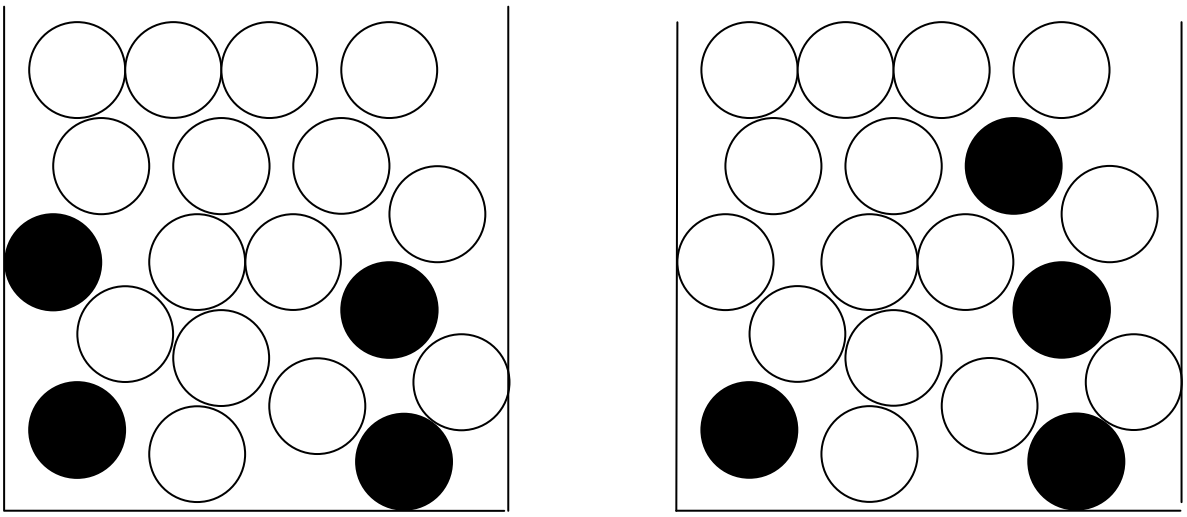
both physically indistinguishable and pertain to one ψ function which is the solution to the Schrödinger equation for our quantum problem; it follows herefrom that within the framework of the stationary problem, the question of the mechanism of atomic motion cannot even be posed.



a) b)

Fig.1. *Two identical pictures of atomic arrangement in the vessel. In the case of distinguishability of atoms, they may be numbered, and the physical state illustrated in Fig. (a) differs from that in Fig. (b). Both of these states without numbers would be physically indistinguishable.*

For the sake of simplicity, we shall the 2D phase space will be represented in what follows. On the other hand, the mass transfer in a liquid may be described in terms of vacancy transfer. Let us consider two configurations differing in positions of vacancies in Fig. 2.



a) b)

Fig. 2. *Two physically distinguishable situations of atomic arrangement in the vessel. They are deduced one from another by moving vacancies over a macroscopic distance (black circles indicate the vacancies). In Fig. (a), the vacancies are distributed more uniformly than in Fig. (b).*

These two configurations are physically recognizable already, because the vacancies at the left are concentrated in one part of the vessel, and the vacancies at the right are clustered in the other part. From this it follows that there must exist be a mechanism for the transition of atoms from one configuration to the other and, this mechanism is associated with atomic motion and, hence, this has a bearing on the superfluidity phenomenon. atomic motion; and thus it is somehow related to the superfluidity phenomenon. What actually happens is that superfluidity manifests itself when the system becomes unbalanced with the resultant atomic motion.

We now consider the physical pictures in Fig. 2 in the configuration 3*N-dimensional (N is the number of atoms in a liquid) space. A point in the configuration space corresponds to each physical state. The presence of a local potential energy minimum gives rise to atomic oscillations (ordinary phonons). This implies that a certain volume (a 3*N-dimensional sphere), rather than a point, in the configuration state corresponds to each physical state (Fig. 3) . The radius of the sphere is not large: it is the atomic oscillation amplitude.

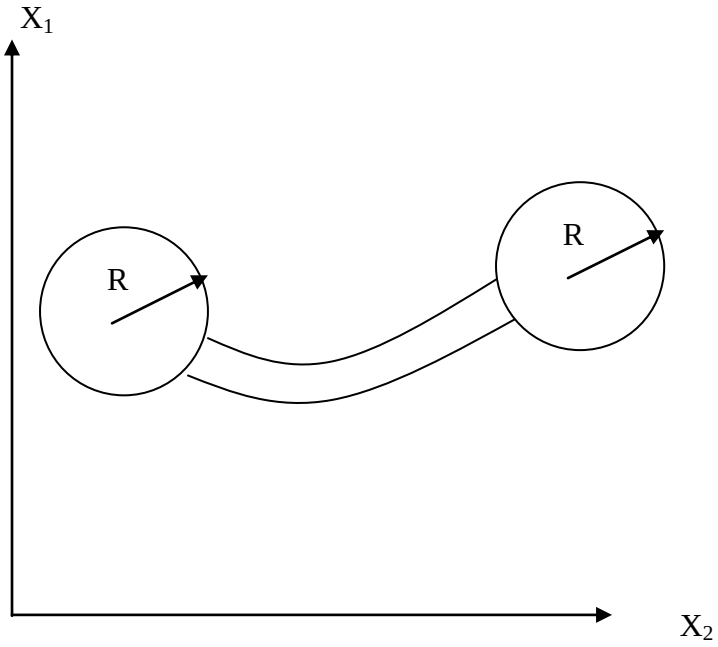


Fig. 3. *The distance between spheres (the sphere's radius R is equal to an average atomic motion due to oscillatory motions) in the configuration space being well over the radius. Each sphere consists of points with the system being there owing to oscillations with reference to a local minimum of potential energy.*

Atoms in liquids are labile and, hence, the system can pass from one point of the configuration space to another. Note that the points corresponding to the atomic configuration in Fig. 2(a) and 2(b) must be far apart, as the vacancies have moved over distances comparable to the size of the vessel. If we draw any line joining two different physical states, we see that its points are outside the spheres and cannot be described by ordinary phonons. The presence of oscillations with respect to a relatively optimal trajectory results in a transformation of the thin fiber exhibiting the transition from one point to another into the cylinder of finite diameter. The displacement over macroscopic distances is only possible with a very labile atom (vacancy) and

without energy exchange with the remaining atoms. But this is possible only in the gas of noninteracting particles.. In the liquid state, motion of some atoms does not seem to be possible; this does contradict the very nature of a condensed state.

On the other hand, there is a mechanism of atomic motion in a liquid which was predicted in investigations using the molecular dynamics method [1]. It implies string-like linear motions of many atoms. The string-like atomic motions observed in the computer experiment comprised, of course, only a few tens of atoms at most. In a classic liquid, because of the presence of large temperature fluctuations, such a wave of successive atomic displacements must be short-lived. This is exactly what is observed in the numerical experiment. If we extrapolate this mechanism to a low-temperature region, we obtain the following situation.

With decreasing temperature, the lower level begins to be filled up, and, since the system retains its mobility, atoms at this level are delocalized. The fact that the energy level of condensate ε_{con} with atoms delocalized in one dimension may lie below the lowest energy level of atoms in an ordinary liquid ε_{liq} can be seen from the expression for this value:

$$\varepsilon_{liq} = \frac{1}{2*m}(P_x^2 + P_y^2 + P_z^2) \geq \frac{1}{2*m}(\frac{\hbar^2}{\Delta X^2} + \frac{\hbar^2}{\Delta Y^2} + \frac{\hbar^2}{\Delta Z^2}), \qquad (2)$$

where ΔX, ΔY, ΔZ is the atom coordinate uncertainty (approximately equal to 0.1 of the interatomic distance) in an ordinary liquid. The dislocation in one of the coordinates causes the energy to attenuate.

The manifestation of superfluidity is atomic motion along linear directions to macroscopic distances in the inertial reference systems (IS). Let us call such states the superfluid strings or the superstrings for brevity. Thus the superfluid ^{4}He seems to be a normal liquid penetrated by superstrings, and their concentration is proportional to the SP density ρ_s. The possible trajectories in the phase space are shown in Fig. 4.

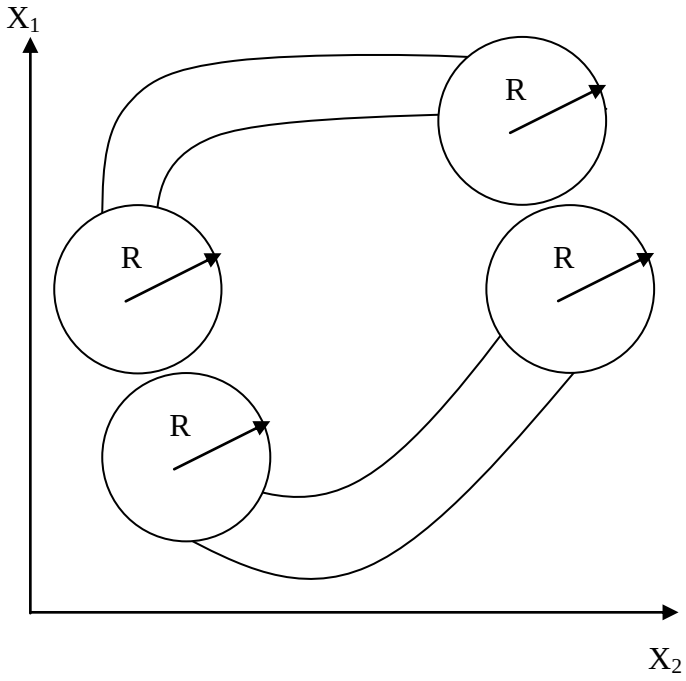


Fig.4. *Adjacent spheres portray the configuration space points resulting from one-particle, usual atomic motions; spaced ones, from atomic motions over macroscopic distances, i.e. superfluid motions. The configuration space regions located within thick fibers are oscillations with respect to a relatively optimal trajectory of soliton motion along the superstirng.*

The proportion of atoms which oscillate routinely (the normal phase) and atoms involved in string-like motion (SP) will be proportional to the volume of corresponding regions in the phase space. Our assumption suggests that the number of superstrings in the differential of volume, dV, is proportional to ρ_s, and they move isotropically in different directions. In this case, it can be shown (reasoning is identical in phonon mechanism, say, to heat conduction [12]) that the sum vector of the superfluid phase velocity is expressed by the formula:

$$\overline{V_s} = -\alpha * grad(\rho_s), \qquad (3)$$

where α is a coefficient from which the condition (1) immediately follows..

It is surprising but there is a mathematical apparatus to describe atomic motions driving the mass transfer in linear objects interacting with the environment, namely the Frenkel-Kontorova model [13]. Importantly, with a nonlinear potential of interatomic interaction and nonlinear interaction with the environment, the ground state may be a chain with a moving!!! soliton. Notice that the motion of the soliton which transfers the mass along a string is the ground state (with minimum energy), not the excited state, i.e. from the state of rest. Therefore, the mass transfer does not imply the transfer of excess (with respect to the ground state) energy and entropy.

3. Corollaries of the assumption about the string-like structure of the superfluid phase

Superstrings must be rectilinear at the microscopic level, because the presence of bendings would point to the momentum or angular momentum exchange with the normal phase. The rectilinearity of strings, the isotropy of their directions and the dependence of their concentration on density ρ_s, which, in turn, may depend on space coordinates, render the fulfillment of the condition (1) possible.

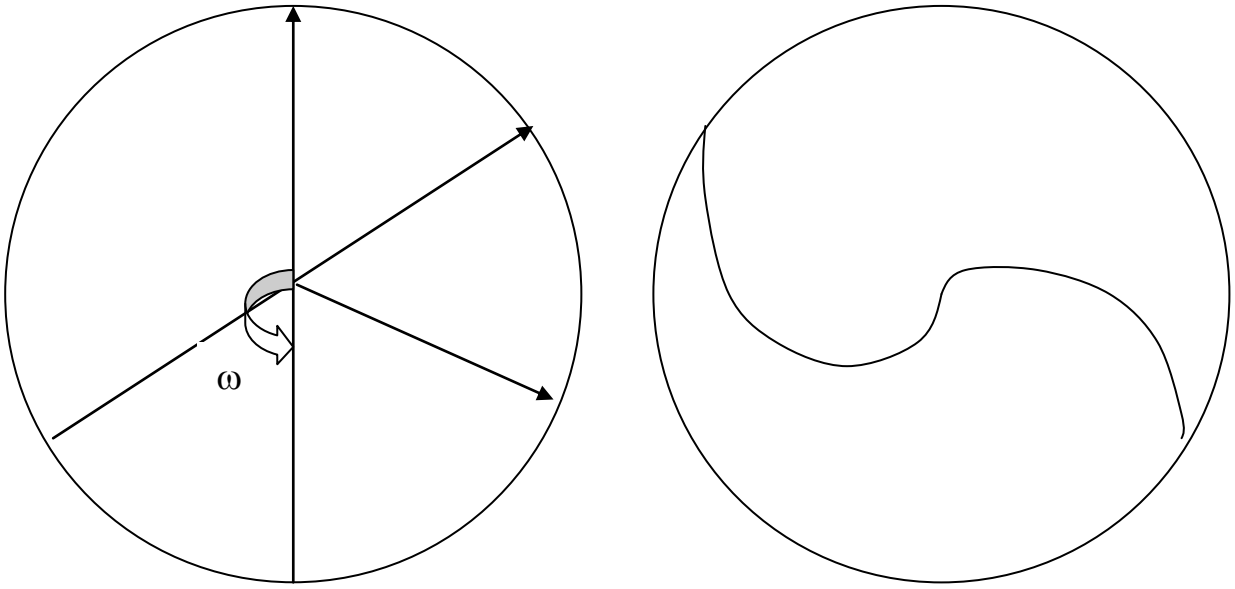
The origination intensity of superstrings in the selected ^{4}He volume depends on local temperature, because the relation between normal density and SP density is a function of temperature [14]. Therefore, with the local nonequilibrium heating of the volume, the number of emerging superstrings will be smaller than that in the ambient ^{4}He volume, and for a while the number of incoming superstrings will exceed that of exiting ones. This will produce a thermo-mechanical effect [11, p.38]: the mass inflow to the heated volume increases. And vice versa, if using superinterstices we succeed only in transferring the mass as superstrings, we will see a mechanocaloric effect: a volume with predominant incoming superstrings cools down.

And the last, most mysterious and intriguing phenomenon is the rotation of ^{4}He which cools down below the λ point (the point of transition to a superfluid state). In the noninertial system

(NS) rotating with the vessel, the observer can observe the following picture: the liquid at rest, when cooling below the λ point, starts rotating. The observed effect might take place only if SP appears in the vessel's center and moves at some speed to the periphery. In this case, the Coriolis force would specify the right direction of the SP rotation. As will be shown later in the text, it is an entirely reasonable assumption. If it is assumed, as done above, that SP has a zero momentum (Bose-Einstein condensate), the origin of its rotation is inexplicable.

4. Thermodynamics of the superfluid phase rotation

We now consider the motion in a plane perpendicular to the rotational vector ω. Let the velocity of soliton motion along a superstring be V_{sol}. In the inertial reference system (IS), superstrings must be rectilinear (Fig. 5a).



a) b)

Fig. 5. *The top view of the cylinder vessel rotating with an angular velocity ω . Fig. a) shows superstring trajectories in the inertial reference system. Fig. b) presents the same trajectories in the rotating reference system.*

In NS, the superstring issuing from the centre of rotation is curved (Fig. 5b), and such a curve is referred to as the Archimedean spiral [15]:

$$r(\theta) = \frac{V_{sol}}{\omega} * \theta \ ,$$

where θ is the polar angle, ω is the angular velocity of rotation, and r is the distance from the center of rotation to the soliton which runs along the string. It can be assumed that the excess (as compared to the rectilinear state) energy ΔE is proportional to the product of the curvature by the

string length. Considering that the curvature is different at each point of the Archimedean spiral, we arrive at the integral:

$$\Delta E = C * \int_0^{\theta_0} \kappa(\theta) * dl(\theta) \qquad \theta_0 = \frac{\omega * d}{2 * V_{sol}} \quad ,$$

where C is a dimensional constant, k(θ) is curvature, dl(θ) is the arc length, and d is the diameter of the vessel with ^{4}He. Substituting the known expressions for curvature and arc length into the dependencies on the θ angle [15] gives:

$$\Delta E = C * \int_0^{\theta_0} \frac{(\theta^2 + \frac{3}{4} * \frac{1}{\theta^2})}{(\theta^2 + \frac{1}{4 * \theta^2})^{\frac{3}{2}}} * (1 + \theta^2)^{\frac{1}{2}} d\theta \approx 12 * C * \theta_0^2 \quad .. \qquad (4)$$
$$\theta_0 \langle\langle 1$$

Thus the excess energy of an isolated superstring at $\theta_0 << 1$ will be proportional to θ_0^2. The number of superstrings is naturally proportional to ρ_s. Hence the free energy expression as used in the theory of second-order phase transitions [16] acquires an additional term:

$$\Delta F = A * \rho_s * \omega^2 \qquad (5)$$

It follows from this formula that the field adjacent to the ρ_s-order parameter is the angular velocity squared and that the phase transition to the superfluid state of rotating helium must become a first-order transition. To find a critical angular velocity ω_k beyond which SP becomes involved in the rotation, it is necessary to calculate a constant **A** from ^{4}He physical parameters or use the qualitative assessments made below.

5. Estimating a critical rotation velocity at which superfluid phase starts to rotate

Experimental evidence [11] shows that when a critical velocity ω_k is exceeded, the SP tangential velocity distribution in the cylinder vessel is described by the equation:

$$V_\theta = \frac{\hbar}{m * r} \quad , \qquad (6)$$

where m is the mass dimension parameter, r is the distance from the center of rotation, and $\hbar$ is the Planck constant. Note that with this velocity–radius dependence as is the case for linear displacements, the condition (1) is satisfied. Hence, condensate atoms can be driven by two mechanisms, and these atomic motions in a liquid were detected via computer simulations [1–8].

The distribution of normal phase velocities during solid (liquid stationary) rotation is described by the formula:

$$V_\theta = \omega * r \quad . \qquad (7)$$

With such a distribution, in the rotating reference system there appears a centrifugal potential:

$$U_u(r) = m * \omega^2 * r^2 \qquad (8)$$

Potential (8) produces an excess pressure in liquid to which SP is very sensitive (disappears altogether at an excess pressure of only 28 atm). We can suggest that the appearance of SP at the λ point is nonuniform in volume: the smallest potential is in the center of the rotating vessel, just where SP starts up. Besides, potential (8) has acquired an important feature [17]: particles (solitons) in this potential move along trajectories of two types only: rectilinear (with zero angular momentum) and closed elliptic (with nonzero angular momentum) (Fig. 6). Quantization of the angular momentum for elliptic trajectories leads to the law of tangential velocity distribution (6), but there is also a nonzero radial V_r component determined from the energy conservation law. Beyond the critical angular velocity, SP starts to move in elliptic trajectories; here the vessel-volume-averaged V_r is zero; the tangential component V_θ is described by formula (6). Tests [18] confirm our viewpoint: the transition between states with a different SP angular momentum is fast (avalanche-like), but not instantaneous, as coherent motion proceeds along an isolated superstring. In a standard interpretation of SP, where the entire rotating volume is a coherent state, there must be no intermediate angular momentum values. Furthermore, noteworthy are the tests [19] which give evidence of the circular motion of ^{4}He atoms, confirming the existence of the superfluid phase at the atomic level, whereas Bose-Einstein condensate requires macroscopic scales.

The angular velocity ω_k is determined by considering the laws of conservation of energy and angular momentum, when elementary excitations are formed, similar to reasoning when determining a critical velocity of SP. We now turn our attention to the rotating reference system in which below the λ point SP rotates and, when exceeding the angular velocity of ω_k rotation, it begins to decelerate. Let M be the angular momentum of the entire SP mass; I, the moment of inertia. A deceleration is accompanied by a decrease of M (the quantum of change is $\hbar$), and an elementary excitation with the energy ε (*p*) occurs:

$$\frac{M^2}{2*I} \to \frac{(M-\hbar)^2}{2*I} + \varepsilon(p) \approx \frac{M^2}{2*I} - \hbar * \omega + \varepsilon(p)\,. \qquad (9)$$

For the SP deceleration process to be energetically effective, the following condition must be satisfied:

$$\hbar * \omega \geq \varepsilon(p)\,.$$

The excitation energy is determined from the condition that the minimum supplementary momentum of a soliton ΔP moving in an elliptic orbit, which touches the edges of the vessel, is:

$$\Delta P * \frac{d}{2} = \hbar \Rightarrow \Delta P = \frac{2*\hbar}{d} \Rightarrow \varepsilon(p) = \frac{\Delta P^2}{2*m} = \frac{2*\hbar^2}{m*d^2}\,. \qquad (10)$$

A qualitative estimate follows from this formula:

$$\omega_к \geq \frac{2*\hbar}{m*d^2} \qquad (11)$$

The estimates made in [9–11] give for ω_k:

$$\omega_к \geq \frac{4*\hbar}{m*d^2}\ln(\frac{d}{2*a}), \qquad (12)$$

where the **a** parameter is of order of the interatomic distance. The discrepancy between our assessment (11) and the commonly accepted assessment (12) lies not only in the difference between numerical coefficients. It is possible to perform an experiment to prove or disprove the validity of our approach. Let ^{4}He rotate in a vessel having the form of two coaxial cylinders: the outer boundary is d_{max} in diameter, and the inner boundary, d_{min}. According to our view, in the cylindrical vessel there is an inner nucleus of radius d_{zero} that is much larger than the interatomic distance (a calculation of this value will be the subject of further investigation), where SP is at rest (Fig. 6).

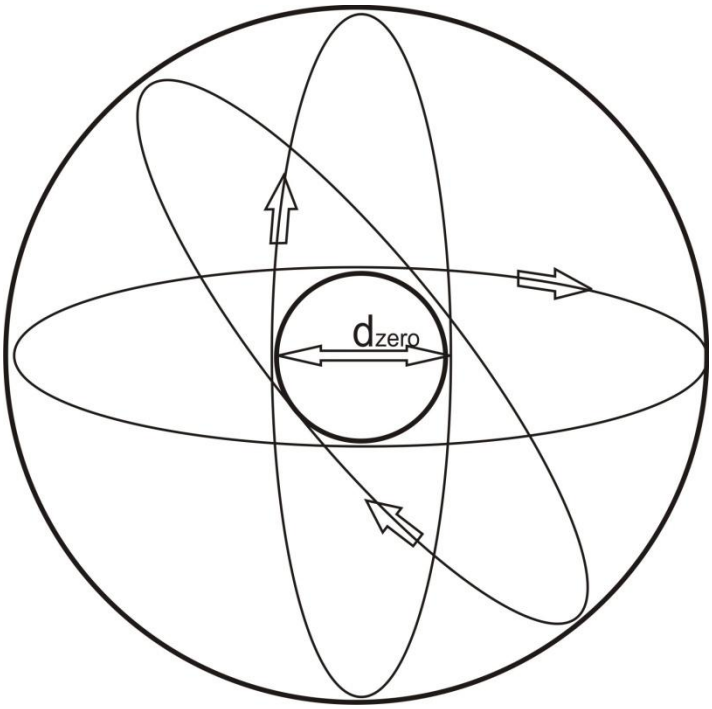


Fig. 6. *Elliptic SP trajectories in the cylinder vessel rotating at an angular velocity exceeding a critical velocity; the arrows indicate the direction of rotation. Also shown here is the outer diameter d_{max} and the inner, interphase, nucleus of radius d_{zero}. The velocity distribution averaged over ellipses of all orientations is described by Formula (6).*

Should d_{min} be very small but much larger than the interatomic distance, then, according to our view, the ω_k values will be independent of d_{min} at $d_{min} < d_{zero}$. In the context of the existing views [9–11], ω_k depends on d_{min} even at the smallest d_{min}.

6. Conclusion

The analysis of the mechanism of atomic motion in liquids suggests the conclusion that, first, most of the motions are of string-like character, and, second, this stringness has only a linear

structure. At decreasing temperature (our unpublished paper relying on a 2D model and other authors' observations) the stringness only increases. With a further decrease in temperature, liquid must solidify or transform to a state in which string-like effects increase. During second-order phase transitions, an order parameter with an almost zero average value has a macroscopic correlation length at the transition point [16]. It is quiet logical to assume that atoms involved in coherent string-like motions make up the superfluid condensate and are the desired order parameter, and this parameter acquires a macroscopic value at the phase transition point. The linear and rectilinear (at the microlevel) character of displacements explains: the fulfillment of equation (1); why the superfluid component is not involved in rotation at low angular velocities; that the field adjacent to the order parameter is the angular velocity squared (5) and, under the assumption of angular momentum quantization, estimates the critical angular velocity (11).

As was emphasized in [7] string-like atomic motion can be observed not only in liquids, but also in a solid, disordered phase. The stringness requires an excess energy and a nonlinear potential of interatomic interaction. This implies that in the amorphous phase, at low temperatures, the mass transfer mechanism may have a string-like character, as is the case with in liquids. Hence, anomalous atomic mobility, known as supersolid, may be seen in the disordered solid ^{4}He [20]. The only state without (by reason of energy) string-like atomic motion is the crystalline state. Experimental data testify that there is no supersolid in annealed samples having good crystal structure [21].

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