Viktor Bobrov, Sergey Trigger* and Daniel Litinski

Universality of the Phonon–Roton Spectrum in Liquids and Superfluidity of ⁴He

Abstract: Based on numerous experimental data on inelastic neutron and X-ray scattering in liquids, we assert that the phonon–roton spectrum of collective excitations, predicted by Landau for superfluid helium, is a universal property of the liquid state. We show that the existence of the roton minimum in the spectrum of collective excitations is caused by the short-range order in liquids. Using the virial theorem, we assume that one more branch of excitations should exist in He II, whose energy spectrum differs from the phonon–roton spectrum. Such excitations are associated with the pole of single-particle Green function, which can have a gap at small values of momenta.

Keywords: Bose-Einstein Condensation; Phonon-Roton Excitations; Single-Particle Excitations; Superfluid Helium; Universality of Collective Excitations.

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1 Introduction

Landau, in his phenomenological theory of superfluidity [1], first introduced the notion of quasiparticles as quantised collective excitations. Based on an analysis of available experimental data on specific heat and second sound, Landau predicted the so-called phonon–roton energy spectrum of such excitations $E_{p-r}(p)$ in superfluid helium, and formulated the criterion of superfluidity disappearance when the flow velocity exceeds a certain critical velocity [1, 2]. To test the Landau hypothesis, Cohen and Feynman [3] proposed to determine the collective excitation spectrum by measuring the position of maxima $E_{max}(p)$ in the dynamic structure factor $S(p, E)$ of liquid.

*Corresponding author: Sergey Trigger, Theoretical Department, Joint Institute for High Temperatures, Russian Academy of Sciences, Moscow 125412, Russia, E-mail: satron@mail.ru
Viktor Bobrov: Joint Institute for High Temperatures, Russian Academy of Sciences, Moscow 125412, Russia
Daniel Litinski: Freie Universität Berlin, Kaiserswerther Str. 16-18, 14195 Berlin, Germany

The function $S(p, E)$ can be experimentally determined by the cross section of inelastic neutron scattering (INS) in liquid, depending on the momentum p and energy E. These ideas were implemented in quite a number of experimental works (see, e.g. [4, 5]), results of which excellently confirmed Landau's assumption on the shape of the spectrum of collective excitations in superfluid helium (see Fig. 1).

Capability of studying the collective dynamics in liquids using INS experiments was later complemented by experiments on inelastic X-ray scattering (IXS) [6, 7].

We emphasise that the experimental data explicitly show the existence of the peaks in the function $S(p, E)$ and the respective curve $E_{p-r}(p)$ of the positions of the maxima in this function for various liquids (as we demonstrate below), not only in superfluid helium. At the same time, the Landau assumption about the existence of rotons, formulated for He II in [1, 2], is still often considered as a specific and typical property of only the superfluid state (see, e.g. [8]).

The function $E_{p-r}(p)$ as the curve describing the position of the maxima in $S(p, E)$ is similar to the phonon–roton excitation curve predicted by Landau in superfluid helium. However, further interpretation of these maxima as some well-defined excitations or quasiparticles was based on the specific theoretical models and the assumptions about the definite shape of the function $S(p, E)$. The existence of quasiparticles means smallness of the respective damping in comparison with the characteristic frequency. At the same time, the maximum of $S(p, E)$ can exist in the case of high damping also. Therefore, it is necessary to distinguish the curve of the maxima in $S(p, E)$ and the statement about the existence of the well-defined quasiparticles in liquids.

Moreover, rigorous theoretical description of collective excitations in liquids is impossible in principle, since interparticle interaction in liquids is strong. Therefore, the existence of quasiparticles and the shape of their spectrum are established in fact by the chosen model for correlation functions of liquids. This choice leads to different interpretations of experimental data.

A general microscopic concept of the Bose condensed systems is based on the existence of a Bose–Einstein condensate (BEC) which is associated, as suggested by
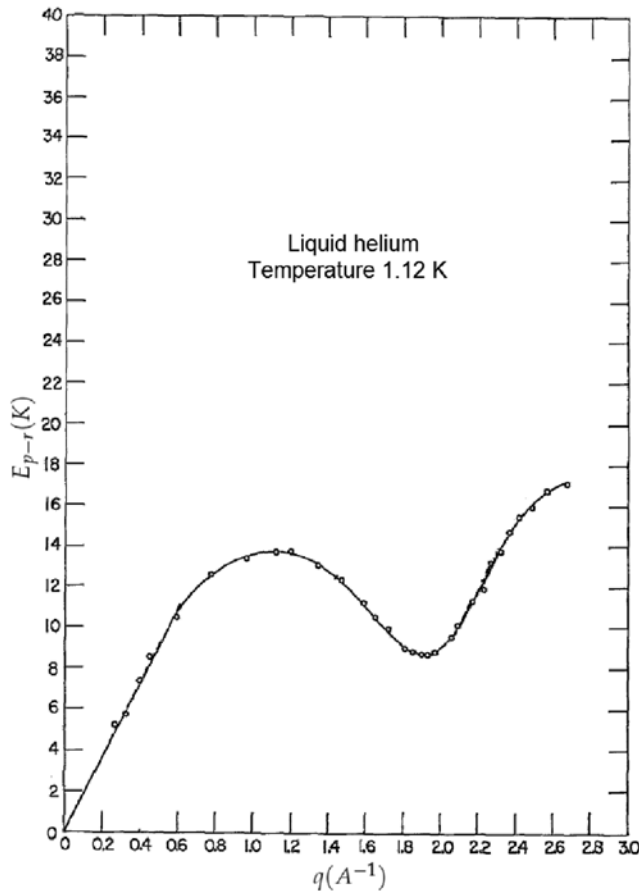
Figure 1: Confirmation of the shape of excitations in superfluid helium, predicted by Landau [2]: typical phonon–roton curve in helium at $T = 1.12$ K [5].

phenomenological theory for He II thermodynamics, based on phonon–roton excitations, can be considered as unexpected success. It should be emphasised that the Landau theory is valid only essentially below the temperature of superfluid transition T_λ. The apparent achievement of the Landau theory [2] is similarity of the spectrum of quasiparticles suggested by Landau and the curve describing the position of maxima of the dynamical structure factor, measured later in INS and IXS experiments.

Nevertheless, we will use below the term phonon–roton branch of excitations, taking into account that the notion of quasiparticles is questionable in the wave vector region where the roton maximum exists.

The paper is organised as follows. In Section 2, we collect the experimental data to show the universality of phonon–roton excitations in liquids. It is especially actual in connection with the recent attempts to qualify these excitations as specific excitations unique for superfluid ^{4}He, which disappear at temperatures T above T_λ [11]. In Section 3, the theoretical background for the universality of phonon–roton excitations in various liquids is formulated. In Section 4, we discuss the possibility of existence of a new branch of excitations, specific only for the superfluid state and disappearing at $T \geq T_\lambda$. We discuss the possible origin of the new branch of excitations as the poles of the single-particle Green function, which can be different from the poles of the retarded "density–density" Green function, which describes phonon–roton collective excitations. The conclusions are presented in Section 5.

Penrose and Onsager [9], with the anomalous spatial behaviour of the equilibrium single-particle density matrix $\rho(\mathbf{r}, \mathbf{r}') \equiv \langle \hat{\Psi}^+(\mathbf{r}) \hat{\Psi}(\mathbf{r}') \rangle$, where $\hat{\Psi}^+(\mathbf{r})$ and $\hat{\Psi}(\mathbf{r})$ are the field operators of creation and annihilation for boson particles, respectively, and angle brackets mean averaging over the Gibbs distribution. In homogeneous and isotropic liquids, the single-particle density matrix has the form $\rho(\mathbf{r}, \mathbf{r}') = \rho(|\mathbf{r} - \mathbf{r}'|)$; hence, the corresponding anomalous spatial behaviour called the off-diagonal long-range order (ODLRO) [10] can be represented as the limit relation

$$\rho(|\mathbf{r} - \mathbf{r}'|)_{|\mathbf{r} - \mathbf{r}'| \to \infty} \to n_0 \neq 0. \tag{1}$$

Here n_0 is the density of boson particles in a BEC. In a normal system BEC is absent and $n_0 = 0$. However, up to now it has failed to give a detailed description of the behaviour of superfluid helium within the ODLRO concept.

In contrast to this concept, the Landau phenomenological theory does not use the notion about the Bose–Einstein condensation. In other words, the Landau

2 Universality of Phonon–Roton Excitations: The Selected Experimental Data

To date, there are experimental data demonstrating the existence of the dynamic structure factor maxima in a wide temperature range for many liquids with various physical properties. Below we present the collection of selected experimental data that seem necessary due to the wide spread opinion, mentioned above, that rotons, as a part of phonon–roton collective excitations, exist only in the superfluid state. The experimental data convincingly show that the phonon–roton spectrum can be considered as a universal characteristic of the liquid state:

(A) Conductive liquids–liquid metals. Experimental data for various liquid metals are given in the reviews [11, 12]. The analogy between the spectra of collective excitations in He II and liquid metals was indicated in [13]. As
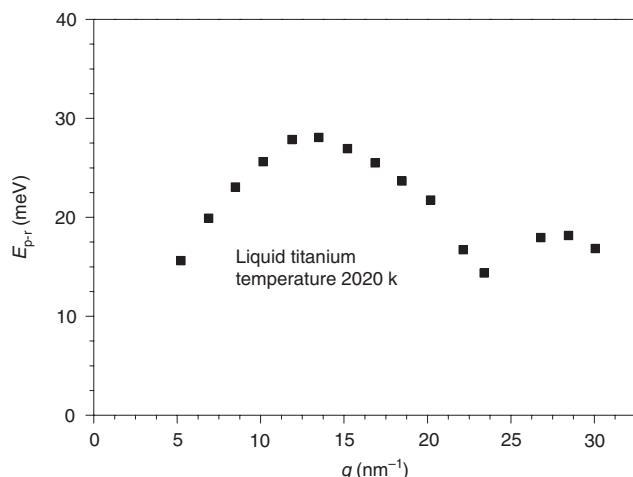
Figure 2: The curve of the position of the dynamical structure factors maxima in liquid titanium [14].

Figure 4: The experimental INS phonon–roton curve for molecular para-hydrogen at temperature 15.7 K. The filled symbols (inverted triangles, diamonds and crosses) depict data measured at different spectrometers and the open circles with a dot show the simulation results [20].

an illustration, we present the results of later experiments for liquid titanium [14] (Fig. 2). Furthermore, a similar excitation spectrum takes place for mercury in the vicinity of the metal–nonmetal transition [15, 16]. These experimental data correspond to the results of calculations by the molecular-dynamics method (see [17, 18] and references therein).

(B) Nonconductive liquids. In [5], the phonon–roton spectrum was experimentally observed in normal liquid helium and the temperature dependence of the roton minimum quantity Δ (Fig. 3) was determined. The analogy between the excitation spectrum in superfluid helium and that in normal liquid was indicated in the experimental work [19].

The corresponding experimental data are also available for molecular para-hydrogen [20] (Fig. 4) and neon [21]. For oxygen, the similar experimental results were
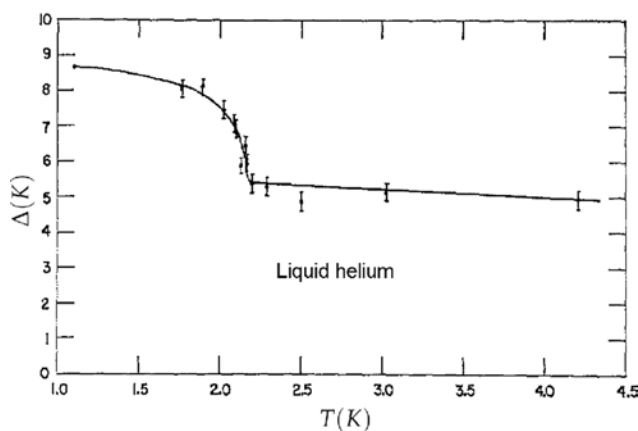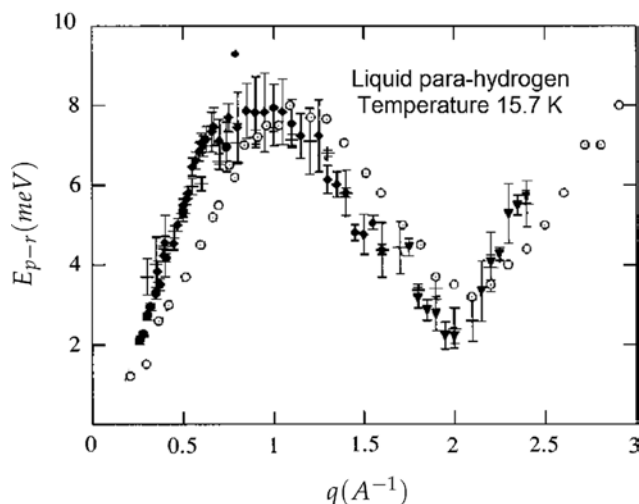

Figure 3: The experimental temperature dependence of the roton gap in superfluid and normal helium [5].

obtained in the supercritical region of thermodynamic parameters [22]. These data validate the assumption (based on the analysis of molecular-dynamic calculations) that the phonon–roton spectrum of collective excitations is the characteristic of dense fluids [23]. The possible existence of two branches of phonon–roton-type collective excitations detected in fluid argon in the supercritical range of parameters should also be noted [24], which confirms the results of numerical calculations (see [25] and references therein) (Fig. 5).

(C) Liquids consisting of several chemical elements. In particular, there are the experimental data on the existence of the phonon–roton spectrum of collective excitations for liquids consisting of very complex molecules of lipid bilayers [26] (Fig. 6) and hydrated β-lactoglobin [27]. The molecular-dynamic calculations confirm the existence of such a branch of excitations in binary liquids [28, 29].

(D) Two-dimensional Fermi liquid. The phonon–roton spectrum of collective excitations was obtained based on INS experiments for a two-dimensional layer of liquid ^{3}He in the normal state [30]. Thus, it was experimentally found that well-defined collective excitations with the momentum larger than $2p_F$ (where p_F is the Fermi momentum) exist in a many-body Fermi system [30]. We should especially emphasise the similarity of the phonon–roton spectra for two-dimensional ^{3}He and bulk superfluid ^{4}He. We should also mention the study of two-dimensional quantum systems by numerical methods (see [31, 32] and references therein).
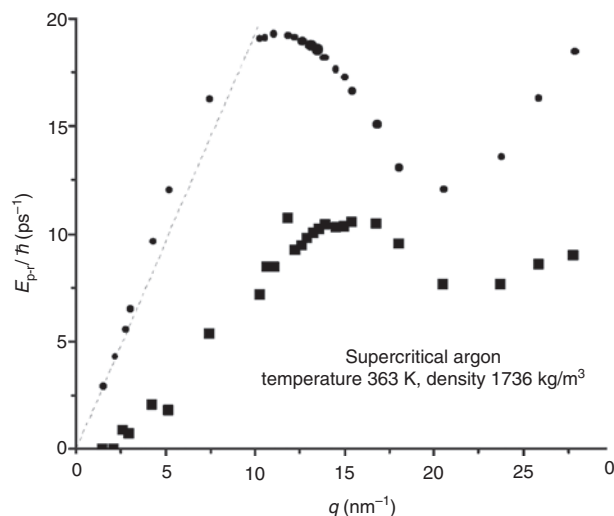
Figure 5: Energy spectra for two types of collective excitations in supercritical *Ar* at temperature *T* = 363 K calculated on the basis of the generalized collective modes approach [24]. Dispersion of acoustic excitations (circles) and non-hydrodynamic heat waves (squares) at a density of 1249 kg/m³. The dashed line corresponds to the hydrodynamic dispersion. In the long-wavelength limit, heat waves do not exist.

3 Universality of the Phonon–Roton Excitations: Theoretical Justification

Let us now turn to theoretical justification of the phonon–roton excitations universality in liquids. First, we note the general feature in the available experimental data for various liquids: the position of the roton minimum p_0 in the spectrum of collective excitations qualitatively corresponds to the position of the first maximum in the static structure factor $S(p)$. The value $S(p)$ can be measured in the experiments on elastic neutron and X-ray scattering and directly related to the dynamic structure factor $S(p, E)$ as

$$nS(p) = \int_{-\infty}^{\infty} \frac{dE}{2\pi\hbar} S(p, E), \qquad (2)$$

where n is the average density of atoms in the liquid under consideration. Sometimes the wave vector $q = p/\hbar$ and frequency $\omega = E/\hbar$ are used in the structure factors
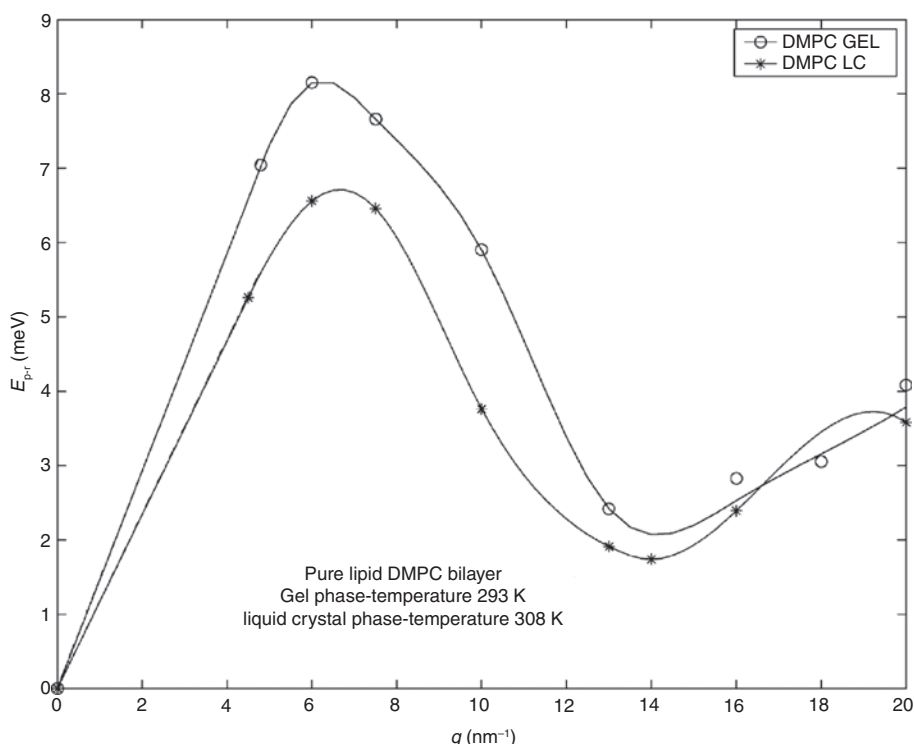

Figure 6: The experimental (IXS) phonon–roton dispersion curves for dimyristoylphosphatidylcholine (DMPC) bilayer at two temperatures, corresponding to the gel and liquid crystal phases of the bilayer. In the gel phase (*T* = 293 K), the hydrocarbon chains are ordered; hence, the phonon frequency and the sound speed are higher. In the liquid crystal phase (*T* = 308 K) the chains are disordered; hence, the phonon frequency and the sound speed are lower. The roton gap at *k* = 14.0 nm⁻¹ is also smaller for the liquid crystal case [26].

as variables. As is known [33], the presence of peaks (maxima) in the static structure factor, the value of which decreases with increasing wave vectors, is associated with the so-called short-range order in liquids. Therefore, it becomes clear that the existence of the roton minimum in the collective excitation spectrum can be a prominent feature of liquids independently of their physical properties, which corresponds to both the experiments and the molecular-dynamic calculations mentioned above. This statement is in qualitative agreement with the known Feynman formula [34] for the phonon–roton spectrum of collective excitations in He II,

$$E_{\text{p-r}}(p) \simeq \frac{p^2}{2mS(p)}. \tag{3}$$

Here m is the atomic mass in the liquid under consideration.

Note that the Feynman formula (3) derivation is based on the analysis of the ground state of the Bose liquid [34]. Therefore, this formula can describe the experiments in superfluid helium only qualitatively. To ensure quantitative agreement with experimental data, relation (3) can be converted to the form $S(p) = p^2/\{m[E_{\text{p-r}}(p) + \Sigma(p)]\}$, where $\Sigma(p)$ is a certain slowly varying function of the momentum p, which appears due to some nonlinear multi-body interactions occuring within a degenerate Bose liquid [35].

Thus, to verify the validity of the statement about the universality of the phonon–roton spectrum for liquids, it is necessary to obtain a relation similar to (3), without taking into account the specifics of the degenerate Bose liquid. Using the exact relation [36]

$$S(p, E) = -\frac{2\hbar}{1 - \exp(-E/T)} \text{Im} \chi(p, E),$$

$$S(p, -E) = \exp(-E/T)S(p, E), \tag{4}$$

we note that the dynamic structure factor $S(p, E)$ is directly related to the "density–density" response function $\chi(p, E)$. Here T is the liquid temperature in energy units.

The "density–density" response function $\chi(p, E)$ is an analytic function in the upper half-plane of complex values E [37], so it is also called the "density–density" retarded Green function [38]. Furthermore, being an analytical function in the upper half-plane of complex E, the response function $\chi(p, E)$ satisfies the Kramers–Kronig relations [37, 38], a direct consequence of which is the exact sum rule (see [39] and references therein).

$$-\frac{2}{\pi} \int_0^\infty dE\, E\, \text{Im} \chi(p, E) = \frac{np^2}{m}. \tag{5}$$

Consequently, the function $\chi(p, E)$ has a pole $\varepsilon(p)$ in the lower half-plane of complex values E,

$$\chi(p, E \to \varepsilon(p)) \to \infty, \quad \varepsilon(p) = E(p) - i\Gamma(p), \quad \Gamma(p) > 0. \tag{6}$$

The response function $\chi(p, E)$ completely defines the spatial and temporal behaviour of the average density of the non-equilibrium liquid in a weak scalar field [36–38]. Thus, if the condition

$$\gamma(p) = \frac{\Gamma(p)}{E_{\text{p-r}}(p)} \ll 1 \tag{7}$$

is fulfilled, the quantity $E(p)$ can be regarded as the quasiparticle energy spectrum $E_{\text{p-r}}(p)$, which corresponds to the collective excitations in liquids. Here $\Gamma(p)$ is the characteristic of the collective excitation damping, which shows the degree of determinacy of the maximum of the dynamic structure factor. The quantity $\hbar/\Gamma(p)$ defines the "lifetime" of corresponding quasiparticles.

The response function $\chi(p, E)$ in the vicinity of the pole $\varepsilon(p)$ (6) can be written as [36]

$$\chi(p, E) = \frac{Z(p)}{E - E_{\text{p-r}}(p) + i\Gamma(p)}, \tag{8}$$

where $Z(p)$ is a certain unknown function of momentum. As condition (7) is satisfied, the imaginary part of the response function $\text{Im} \chi(p, E)$ in the vicinity of the pole $\varepsilon(p)$ can be written in terms of the Dirac δ-function,

$$\text{Im} \chi(p, E) = \frac{\Gamma(p)Z(p)}{(E - E_{\text{p-r}}(p))^2 + \Gamma^2(p)} \approx -\pi Z(p)\delta(E - E_{\text{p-r}}(p)). \tag{9}$$

Let the representation (9) be valid for any momentum. Then, taking into account (4) and substituting (9) into (2) and (5), we arrive at the following result [40, 41]:

$$S(p) \simeq \frac{p^2}{2mE_{\text{p-r}}(p)} \coth\left(\frac{E_{\text{p-r}}(p)}{2T}\right). \tag{10}$$

This relation is the generalisation of the result obtained by Feynman and Cohen [42] for the structure factor at a finite temperature by using the collective excitation spectrum in the long wavelength limit. As is known, the limit relation

$$\lim_{p \to 0} S(p) = n\kappa_T T \tag{11}$$

is fulfilled for any liquid. If the condition $E_{\text{p-r}}(p) \ll T$ is valid, it directly followed from (10) and (11) that the energy spectrum of collective excitations $E_{\text{p-r}}(p)$ at small momenta $(p > 0)$ corresponds to phonons,

$$E_{\text{p-r}}(p \to 0) = \hbar s_T p, \qquad (12)$$

where s_T is the isothermal speed of sound and κ_T is the isothermal compressibility of liquid. It should be emphasised that, according to (10)–(12), the Feynman formula (3) at finite temperatures is not valid in the region of extremely small momenta [40, 43]. In turn, if the condition $E_{\text{p-r}}(p) \gg T$ is true, (10) corresponds to the Feynman formula (3), as required.

In other words, the result of using formula (10) at small momenta depends on the order of limit transitions: $T \to 0$, $p \to 0$ or $p \to 0$, $T \to 0$. In the former case, the result is the Feynman formula (3); in the latter case, the limit result corresponds to (11). Only the second limit transition has a physical sense at small momenta, since temperature is a physical parameter of the system, which has a well-defined nonzero value. Therefore, the transition to the limit $T \to 0$ means $T/E \to 0$, where E is the characteristic energy. In this case, this value is the collective excitation energy.

We note that numerical calculations based on relation (10) confirm the existence of the "maxon" maximum, as well as the roton minimum in the collective excitation spectrum for various model systems (see [41] and references therein). However, the quantitative description of the collective excitation spectrum cannot be obtained in such a way.

We note that the results presented in this section are valid for both Fermi liquid and Bose liquid. These results are derived under the sole assumption that only one type of the collective quasiparticles exists in the liquid under consideration (9). This means that we can apply the Feynman formula (3) to a qualitative analysis of the collective excitation spectrum in normal liquids when the condition $E_{\text{p-r}}(p) \gg T$ is valid. Otherwise, we should use the general relation (10).

Therefore, if there are well-defined collective excitations in the range of momenta (wave vectors) corresponding to the position of the first maximum of the static structure factor in liquid, the excitation spectrum in this range of momenta is characterised by a roton minimum.

As a result, we arrive at the conclusion that, at least in the region of not very high temperatures, the phonon–roton spectrum of excitations is a universal property of the liquid state. This universality is connected with the short-range order in liquid, but is not a feature of the superfluid state. In other words, the phonon–roton spectra of collective excitations in liquids are caused to a greater extent by the interaction of particles, rather than by quantum statistics effects [41, 44, 45].

4 New Excitations in He II

Thus, the collective excitations with the phonon–roton spectrum are not directly related to the superfluidity phenomenon. In this regard, we pay attention that the Landau superfluidity criterion [1] is in fact the criterion of the superfluidity disappearance [46]. Otherwise, taking into account the existence of phonons and rotons in most liquids, we would arrive at the conclusion about the existence of superfluidity in different liquids until the flow velocity exceeds the corresponding critical velocity. In other words, the superfluidity criterion is based on the consideration of already existing superfluid flow in He II.

Furthermore, the statement that rotons are not directly related to the superfluidity phenomenon in He II is confirmed by the known fact that the critical velocity in He II, which is calculated by using the roton minimum, is higher than that observed in experiments by orders of magnitude [47, 48].

For this reason, we pay attention that Landau [1] initially considered two types of collective excitations in He II, namely acoustic excitations which are in any liquid and vortex excitations (rotons) that are specific for superfluid helium. In addition, Landau suggested that the energy spectrum of the vortex excitations has a gap at small momenta ($p \to 0$). Then, in the calculation of thermodynamic properties and velocity of second sound in superfluid helium, Landau [2] combined these two branches of excitations into a single branch of longitudinal phonon–roton excitations. In this case, the roton spectral region should no longer be associated with vortex motion and the roton minimum in the spectrum is at a nonzero momentum [49].

Feynman [50] implemented the primary Landau's idea about the existence of vortices as some quantised excitations inherent only to the superfluid state. The quantised vortices are actually observed experimentally in He II (see [50] and references therein). As a result, the superfluidity disappearance during He II motion in capillaries is currently associated with generation of extensive quantum vortices or closed vortex filaments (loops, rings), which, in particular, allows description of the experimental dependence of the critical velocity on the size of capillary holes [51–53]. However, the existence of vortices at temperatures above the transition temperature T_Λ in the superfluid state has not been resolved experimentally. In addition, in the presence of vortices, He II should be considered as an inhomogeneous nonequilibrium liquid. To describe inhomogeneous nonequilibrium Bose fluids, the Gross–Pitaevskii equation is commonly used (for details, see [54]), as well as its modifications, which take into

account the interaction between particles (see [55] and references therein).

At the same time, according to the foregoing analysis, the dynamic structure factor $S(p, E)$ and the response function $\chi(p, E)$ are the equilibrium correlation functions. This means that the phonon–roton spectrum of collective excitations characterises the equilibrium state of liquids. In [2] Landau used this spectrum to calculate the thermodynamic functions of superfluid helium.

Within statistical thermodynamics, the transition of liquid helium from the normal state to the superfluid state is a second-order phase transition. According to the existing ideas such a phase transition is associated with the appearance of the BEC in liquid helium or the superfluid component according to the Landau theory [1]. We believe that the concept of the superfluid component only makes sense to describe the phenomenon of superfluidity as such. Therefore, the density of the superfluid component is a function of the BEC density.

Since the phonon–roton collective excitations are not a feature of the superfluid phase of liquid helium, we assume that there are some other specific excitations in He II, which are characteristic exactly of the superfluid state. They can exist only in superfluid helium in addition to phonon–roton excitations. These excitations should have the temperature dependence leading to their disappearance at temperatures above the phase transition temperature T_λ. This hypothesis ensues from the Landau idea about the specific role of excitations in the superfluid state, which are also crucial for the description of thermodynamic properties. This statement is also consistent with the use of specific elementary excitations (Cooper pairs) to describe the superconducting state. We will refer to these new excitations as "helons" to distinguish them from rotons, phonon–roton excitations, and quantised vortex excitations in superfluid helium [56].

To verify the possible existence of helons in the Bose liquid, let us consider the Gibbs thermodynamic potential $\Omega(T, \mu, V)$, where μ is the chemical potential of liquid which occupies macroscopic volume V at temperature T. Below we describe the detailed procedure for calculating the thermodynamic functions of the quantum system to demonstrate how the accurate transition to the thermodynamic limit leads to the opportunity to take into account the BEC without using anomalous averages. We should take into account that the averaging with the Gibbs distribution corresponds to the state of thermodynamic equilibrium only after transition to the thermodynamic limit

$$\langle \hat{N} \rangle \to \infty, \quad V \to \infty, \quad n = \langle \hat{N} \rangle / V = \text{const.} \tag{13}$$

Here $\hat{N} = \int_V d^3r \hat{\Psi}^+(\mathbf{r}) \hat{\Psi}(\mathbf{r})$ is the operator of the total number of particles in volume V. Therefore, to calculate average values, the system should be initially considered in a very large (macroscopic), but finite volume V, and then the transition to the thermodynamic limit should be performed [57]. The chemical potential and temperature are independent intensive thermodynamic parameters in the grand canonical Gibbs distribution. This means that the transition to the thermodynamic limit (13) is equivalent to the limit $V \to \infty$. Therefore, the liquid pressure is given by

$$P(T, \mu) = -\lim_{V \to \infty} \frac{\Omega(T, \mu, V)}{V}. \tag{14}$$

If the function $P(T, \mu)$ is known, all thermodynamic functions of the liquid are completely determined. In particular, the liquid density can be found from the equation $n(T, \mu) = (\partial P / \partial \mu)_T$. The use of the virial theorem for calculating the pressure yields the expression (see, e.g. [38])

$$P(T, \mu) = \lim_{V \to \infty} \frac{2 \langle K \rangle^{(V)}}{3V} - \lim_{V \to \infty} \frac{\langle \mathbf{r} \cdot \nabla U(\mathbf{r}) \rangle^{(V)}}{3V}. \tag{15}$$

Here $\langle K \rangle^{(V)}$ is the average kinetic energy, index (V) indicates that the system is in a very large but finite volume V, $\langle \mathbf{r} \cdot \nabla U(\mathbf{r}) \rangle^{(V)}$ is the so-called average virial of the potential energy,

$$\langle \mathbf{r} \cdot \nabla U(\mathbf{r}) \rangle^{(V)} = \frac{n^2}{2} \int_V d^3r (\mathbf{r} \cdot \nabla U(\mathbf{r})) g^{(V)}(\mathbf{r}). \tag{16}$$

Here $U(r)$ is the pair potential of interaction between particles, and $g^{(V)}(\mathbf{r})$ is the pair correlation function of liquid. The angle brackets in this paper mean averaging with the grand canonical Gibbs distribution.

Since the static structure factor $S(p)$ is uniquely determined by the pair correlation function

$$S(p) = 1 + n \int d^3r \exp(i(\mathbf{pr})/\hbar)[g(r) - 1],$$
$$g(\mathbf{r}) = \lim_{V \to \infty} g^{(V)}(\mathbf{r}), \tag{17}$$

and taking into account (11) and (16), it is easy to verify that

$$\frac{\langle \mathbf{r} \cdot \nabla U(\mathbf{r}) \rangle^{(V)}}{3V} = -\frac{n^2 u(0)}{2}$$
$$- \frac{n}{6} \int \frac{d^3p}{(2\pi\hbar)^3} [3u(p) + (\mathbf{p} \cdot \nabla u(p))](S(p) - 1), \tag{18}$$

$$u(p) = \int d^3r \exp(i(\mathbf{pr})/\hbar) U(r). \tag{19}$$

We should emphasise that the static structure factor of superfluid helium has no experimentally observed features in comparison with $S(p)$ in normal liquid helium. Therefore, relation (18) is valid for any one-component liquid at arbitrary thermodynamic parameters, including the temperature region $T < T_\Lambda$. As a result, taking into account relations (10) and (18), we arrive at the conclusion that the phonon–roton spectrum of collective excitations affects the thermodynamic properties of superfluid helium through the average virial of the potential energy only qualitatively.

Now let us consider the average kinetic energy of liquid in a macroscopic volume V, which can be written as

$$\langle K \rangle^{(V)} = \sum_{\mathbf{p} \neq 0} \frac{p^2}{2m} f^{(V)}(\mathbf{p}), \quad f^{(V)}(\mathbf{p}) = \langle a_p^+ a_p \rangle^{(V)}, \tag{20}$$

where $f^{(V)}(\mathbf{p})$ is the average occupation number of particles with momentum $\mathbf{p}$ (or the single-particle distribution function over momenta), and $\hat{a}_\mathbf{p}^+$ and $\hat{a}_\mathbf{p}$ are the creation and annihilation operators in this state, respectively. Without loss of generality, we further assume that particles have zero spin.

Since the operators $\hat{a}_\mathbf{p}^+$ and $\hat{a}_\mathbf{p}$ are directly related to the field operators as

$$\hat{\Psi}^+(\mathbf{r}) = \frac{1}{\sqrt{V}} \sum_\mathbf{p} \exp(-i(\mathbf{pr})/\hbar) \hat{a}_\mathbf{p}^+,$$

$$\hat{\Psi}(\mathbf{r}) = \frac{1}{\sqrt{V}} \sum_\mathbf{p} \exp(i(\mathbf{pr})/\hbar) \hat{a}_\mathbf{p}, \tag{21}$$

it is easy to verify that the single-particle density matrix of the homogeneous liquid in volume V has the form

$$\rho(|\mathbf{r} - \mathbf{r}'|) = \frac{1}{V} \sum_\mathbf{p} f^{(V)}(\mathbf{p}) \exp(i\mathbf{p} \cdot (\mathbf{r} - \mathbf{r}')/\hbar). \tag{22}$$

Substituting (22) into limit equality (1), we find the relation for the average density of the number of particles in BEC in the homogeneous system

$$n_0 = \frac{f^{(V)}(\mathbf{p}=0)}{V}. \tag{23}$$

This means that the average occupation number $\langle \hat{N}_0 \rangle^{(V)} = \langle \hat{a}_0^+ \hat{a}_0 \rangle^{(V)}$ with zero momentum is a macroscopic quantity defining BEC in a liquid which is characterised by the ODLRO.

As a result, after the transition to the thermodynamic limit, the distribution function over momenta for a liquid in the presence of BEC has the form

$$f(\mathbf{p}) = \langle \hat{N}_0 \rangle \delta_{\mathbf{p},0} + f^{(\text{over})}(p)[1 - \delta_{\mathbf{p},0}], \tag{24}$$

where $f^{(\text{over})}(p)$ is the single particle distribution function for particles in the "overcondensate" state with nonzero momentum $\mathbf{p} \neq 0$. In this case, the average density of the number of particles $n = \langle \hat{N} \rangle / V$ is given by

$$n = \lim_{V \to \infty} \frac{1}{V} \sum_\mathbf{p} f^{(V)}(\mathbf{p}) = n_0 + n^{(\text{over})} \tag{25}$$

$$n^{(\text{over})} = \lim_{V \to \infty} \frac{1}{V} \sum_{\mathbf{p} \neq 0} f^{(V)}(\mathbf{p}) = \int \frac{d^3 p}{(2\pi\hbar)^3} f^{(\text{over})}(p), \tag{26}$$

where $n^{(\text{over})}$ is the density of the number of particles in the overcondensate states. Thus, for calculating the average values corresponding to the thermodynamic equilibrium in liquid in the presence of BEC, it is necessary to consider the initial system in a very large, but finite volume V. Then, after separating the singular terms corresponding to the macroscopic number of particles in BEC, the transition to the thermodynamic limit should be performed. A similar statement takes place when considering inhomogeneous liquid with a BEC [58].

It is evident that the average kinetic energy of a Bose liquid is completely determined by the single-particle distribution function of overcondensate particles at temperatures $T < T_\lambda$. Therefore, according to (20) and (24), we obtain

$$\lim_{V \to \infty} \frac{2\langle K \rangle^{(V)}}{3V} = \frac{2}{3} \int \frac{d^3 p}{(2\pi\hbar)^3} \frac{p^2}{2m} f^{(\text{over})}(p). \tag{27}$$

In the theory of Bose liquids, to calculate the function $f^{(\text{over})}(p)$, the explicit relation is commonly used (see, e.g. [59])

$$f^{(\text{over})}(p) = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{dE}{\exp(E/T) - 1} \text{Im} G(p, E). \tag{28}$$

where $G(p, E)$ is the single-particle retarded Green function for overcondensate states.

As the response function $\chi(p, E)$, the Green function $G(p, E)$ is an analytic function in the upper half-plane of complex values E. Consequently, the function $G(p, E)$ has a pole $\varepsilon_{\text{sp}}(p) = E_{\text{sp}}(p) - i\Gamma_{\text{sp}}(p)$ $(\Gamma_{\text{sp}}(p) > 0)$ in the lower half-plane of complex values E. Thus, if the condition $\Gamma_{\text{sp}}(p) \ll E_{\text{sp}}(p)$ is fulfilled, the quantity $E_{\text{sp}}(p)$ can be regarded as the quasiparticle energy spectrum corresponding to single-particle excitations in liquids. The quantity $\hbar/\Gamma_{\text{sp}}(p) > 0$ defines the "lifetime" of corresponding quasiparticles.

As a result, we conclude that, in addition to the phonon–roton spectrum $E_{\text{p-r}}(p)$ of collective excitations, the

thermodynamic properties of liquid depend on the spectrum $E_{sp}(p)$ of single-particle excitations. This naturally raises the question of the relation between the spectra of quasiparticles $E_{p-r}(p)$ and $E_{sp}(p)$ [60].

These spectra are different in normal liquids (see, e.g. [37]). This difference is caused, in particular, by the physical nature of appropriate quasiparticles. Single-particle excitations describes the "statistical" behaviour of a real particle in a liquid, in contrast to the collective excitations with the phonon−roton spectrum. However, these spectra are assumed to be identical in most papers, when considering the degenerate Bose liquid (for details, see [61]). This statement is entirely based on the Bogolubov assumption [62] that the operators $\hat{a}_0^+$ and $\hat{a}_0$ are the C-numbers,

$$\hat{a}_0^+ = (\langle \hat{N}_0 \rangle)^{1/2}, \quad \hat{a}_0 = (\langle \hat{N}_0 \rangle)^{1/2}. \tag{29}$$

From (29), the existence of the so-called anomalous averages follows (for detail, see [36, 61]). Furthermore, if the hypothesis (29) is valid, the spectrum of single-particle excitations has no gap in the region of small momenta. In this case, at $p \to 0$, the quantity $E_{sp}(p)$ has a form similar to (12) [63].

However, the validity of the hypothesis (29) cannot be proved mathematically correct. At the same time, there are considerable doubts as to whether the statement (29) can be used to describe degenerate Bose liquid [64–70]. In particular, on the basis of (29), another model Hamiltonian is used instead of the original Hamiltonian for the liquid (for detail, see [71]). We note that the presence of ODLRO (1) follows from (29), while the opposite statement is not true.

The alternative approach to describe degenerate Bose liquid, based on the calculation of the thermodynamic functions of liquid in the finite macroscopic volume, is introduced above. The following transition to the thermodynamic limit, taking into account the macroscopic quantity of particles with zero momentum, leads to the self-consistent theory of systems with a BEC. In implementing this approach, we can use the standard methods of the perturbation theory diagram technique [72].

In this case, within the self-consistent Hartree−Fock approximation, it can be shown that the spectrum of single-particle excitations has a gap in the region of small momenta [66] (see also [68–70] for details). The value of the gap is determined by the particle density in BEC. The gap vanishes if we neglect the interaction between particles, or when BEC is absent.

Thus, we can consider the helons as single-particle excitations in liquid in the presence of BEC. For this reason, the helons cannot be observed in INS and IXS experiments. The prominent feature of helons is their disappearance as quasiparticles during the transition of
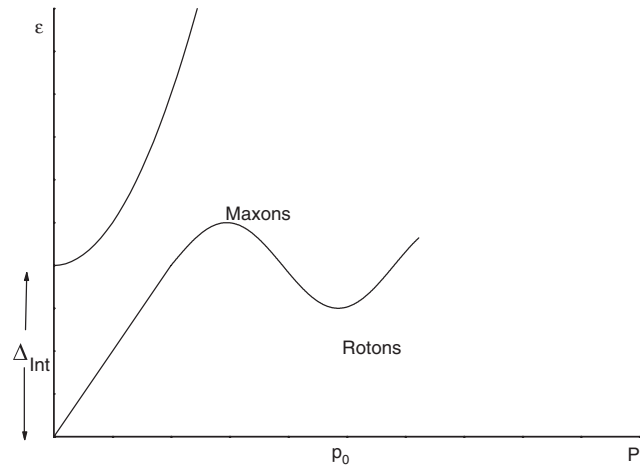

Figure 7: Phonon−roton spectrum and the expected spectrum of helons as the single-particle excitations with the gap $\Delta_{int} \equiv \Delta^{(h)}(T)$ depending on interaction and temperature.

liquid with BEC to the normal state. Given that the liquid is homogeneous and isotropic, without loss of generality, we can assume that the energy spectrum of helons can be presented as $E_h(p) = \Delta^{(h)} + p^2/2m^{(h)}$, where $\Delta^{(h)}(T)$ is the helon gap and $m^{(h)}$ is the corresponding effective mass (see Fig. 7). This spectrum is similar to the roton spectrum first proposed by Landau [1]. The fundamental difference of such quasiparticles from the Landau rotons [1] is that the gap $\Delta^{(h)}(T)$ is a function of temperature, in particular, $\Delta^{(h)}(T) = 0$ at $T > T_\lambda$. Moreover, in the presence of BEC, helons appear only as a result of the interparticle interaction. Within the Landau approach to the thermodynamics of He II [1, 2], the existence of helons gives explanation of the heat capacity singularity at $T \to T_\lambda$ in He II [56].

5 Conclusions

The well-known Hugenholtz and Pines theorem [63] in the absence of a gap in the single-particle excitations spectrum at small momenta is based in principle on the formalism of anomalous averages to describe BEC (see [46] and references therein). Therefore, as shown in [71], there is no limit for the existence of the gap in the single-particle excitation spectrum at small momenta if the anomalous averages formalism is not used for describing the quantum liquid in the presence of BEC.

An alternative approach for describing the quantum liquid in the presence of BEC without using the formalism of anomalous averages may be based on applying the conventional diagram technique of the perturbation theory to an equilibrium system in a large but finite volume [72] and

on the existence of the ODLRO (1) in the single-particle density matrix [9, 10]. Such an approach allows the self-consistent description of BEC by the accurate transition to the thermodynamic limit [73].

However, unlike the spectrum of collective excitations, which is determined by the "density–density" response function poles, the spectrum of single-particle excitations associated with the single-particle retarded Green's functions poles cannot be observed in INS and IXS experiments.

Theoretical consideration in this paper based on the accepted model of He II as a system of interacting neutral particles (see, e.g. [61, 62]). The models related to Coulomb interaction between particles, based on one-component charged bosons (see, e.g. [74]) or two-component (electron-nuclear) boson plasma (see, e.g. [75]) are outside of the considered problems and need a special discussion.

Summarising the above consideration, we can make the following conclusions.

(i) The phonon–roton spectrum corresponding to positions of maxima in the dynamic structure factor is a universal property of liquids.

(ii) Collective excitations with the phonon–roton spectrum are not directly related to the superfluidity phenomenon.

(iii) In the Bose liquid with BEC, the existence of single-particle excitations which can have a gap in the energy spectrum at small momenta is possible.

(iv) Single-particle excitations cannot be observed in INS and IXS experiments. To answer the question about the existence of such excitations, the methodology of an appropriate experiment should be developed.

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