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## РАВНОВЕСИЕ БЭК-КВАНТОВЫЙ ГАЗ И СТРУКТУРА H<sub>2</sub>O ЖИДКОСТИ

*Предложена концепция проявления бозе-эйнштейновского конденсата (БЭК) в спектре, полученном как геометрическое среднее двух масс-спектров, один из которых соответствует частицам при температуре появления БЭК, а другой — равновесию квантовый газ-конденсат на уровне энергии идеального квантового газа по Эйнштейну (1) или тепловой длины волны (2). Масс-спектры равновесия БЭК-квантовый газ H<sub>2</sub>O на линии насыщения жидкости (m) моделируются системами из двух и более кластеров, содержащих резонансы с  $N = 2-12$ . Используя m, удается успешно интерпретировать экстремумы температурной функции конфигурационной теплоемкости тождественных частиц и рассчитать межмолекулярные расстояния и другие характеристики структуры H<sub>2</sub>O по дифракции и интерференции волн де Бройля: в свойствах и структуре метастабильной H<sub>2</sub>O жидкости обнаруживается проявление БЭК вплоть до критической температуры.*

**Ключевые слова:** H<sub>2</sub>O, жидкая фаза, равновесие БЭК-квантовый газ, квантовые волновые масс-спектры, резонансы Ефимова с  $N=2-12$ , квантовый газ тепловой длины волны, квантовый газ по Эйнштейну, масс-спектры и экстремумы теплоемкости; бозонные пики тождественных частиц; дифракция и интерференция волн де Бройля; межмолекулярные расстояния, тетраэдрическая, гексагональная и пентагональная координация.

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## THE BEC-QUANTUM GAS EQUILIBRIUM AND THE STRUCTURE OF H<sub>2</sub>O LIQUID

*The concept of a bose-einstein condensate (BEC) displaying in a spectrum received as of geometrical mean of two mass spectra, one of which corresponds to particles at temperature of BEC occurrence, and another corresponds to equilibrium ideal quantum gas by Einstein (1) or with thermal wavelength (2) is offered. Spectra of H<sub>2</sub>O liquid (m) are modulated by systems of two or more clusters (atoms, "resonances" with  $N = 2-12$ ). Using m, the temperature function extremes of configuration heat capacity of identical particles are successfully interpreted and the intermolecular distances and other characteristics of structure H<sub>2</sub>O in the phenomenon of diffraction and interference of de Broglie waves are designed. It is revealed, that BEC is shown in properties and structure of a metastable and stable H<sub>2</sub>O liquid down to critical temperature.*

**Keywords:** H<sub>2</sub>O, liquid phase, BEC-ideal quantum gas equilibrium, quantum wave mass spectra, Efimov resonance at  $N = 2-12$ ; thermal wavelength quantum gas, Einstein quantum gas, mass spectra and extremums of heat capacity; identical particles heat capacity boson peaks; de Broglie waves diffraction and interference; tetrahedral, hexagonal and pentagonal coordination.

The Einstein predicted transition of a substance from a gas condition into a Bose-Einstein condensate (BEC) [10; 13] takes place in ultracold gases at temperatures  $10^{-6} - 10^{-9}$  K [1; 8; 16]. The influence of Efimov resonances [12; 17] in a cluster condition on recombination speed of the cold gases Bose-Einstein condensate is being investigated [24]. The BEC becomes apparent within the superfluidity phenomena of liquid helium at 2.17 K [7; 15; 18]. It also appears in the superconductivity of solid phase [18], which takes place at temperatures up to and above 135 K [19].

Particle mass, which corresponds to the critical temperature of the BEC formation ( $T_c$ ) [1], has been estimated for liquid ethane [23]. In the case where ( $T_c$ ) temperatures coincide with the temperature ( $T$ ), the results do not contradict with the masses of particles involved in the quantum

gas — condensate reversible transitions. Moreover, at certain conditions the quantitative ratio can be found for compared masses [23].

Within the framework of reversible transitions Einstein quantum gas — condensate model [13] the appearance of proton pairs is revealed in liquid H<sub>2</sub>O [22]. The temperature function extremes of heat capacity of liquid ethane are successfully interpreted [23]. These can be a basis for application of the BEC concept towards properties and structure of liquids. H<sub>2</sub>O as most investigated [4-5; 9; 11; 14; 20-22; 25; 26] and full of puzzles [9; 22; 25; 26] liquid is suitable for application of techniques and equations determined within the framework of such concept.

### The BEC — quantum gas equilibrium concept

The nature of identical particles was found to appear in configuration fluctuations accompanied by reversible transitions quantum gas — condensate [22-23]. The mass of a cluster, containing  $n$  resonantly but not chemically interacting particles, is used as a criterion to identify particles and to evaluate possible formation of Efimov resonances. The coherent movement of identical particles results in an increase of a cluster mass. Two or more interacting clusters constitute a system. The total mass of a system is smaller than the mass of a light cluster, and it decreases with the increasing number of clusters, which could have the same mass. The mass of particles participating in considered processes depends on the energy of quantum gas equilibrium. The temperature function of the mass of particles is called mass — spectrum [23]. There are four levels of energy used for gradation of quantum gas and mass — spectra ( $m_1, m_2, m_3, m_4$  [22–23]). The mass of particles at the temperature of BEC occurrence ( $T_c$ , [1]) are selected in a separate spectrum ( $m_{11}$ ) [23]. Furthermore, mass-spectra were found to transition from one to another. Such transitions could be caused by interactions between clusters as well as with particles of an environment [23]. These interactions were classified as an “interaction of clusters” [24] within the mass-spectrum; however, the interaction between mass-spectra was not considered and, accordingly, the results of such interaction were not discussed.

In this work the possibility of mass — spectra interaction is proposed. The mass of liquid particles, which is given as a result of such interaction, coincides with the geometric mean of interacting masses. The use of geometric mean is necessary to account for the sum of masses as well as for their inverse values. There is a special case with two interacting mass-spectra: when one ( $m_{11}$ ) is the mass-spectrum of particles at the temperature of BEC occurrence, and the other one corresponds to the ideal quantum gas equilibrium ( $m_1, m_2, m_3, m_4$ ). In this work, such case will be called *the BEC-quantum gas equilibrium*. In the indication of mass the indexes of initial mass-spectra will be taken into account, for example,  $m_{411} = (m_4 \cdot m_{11})^{1/2}$ ,  $m_{211} = (m_2 \cdot m_{11})^{1/2}$ .

### Formalism of the mass of particles computation in mass-spectra

The *mass of particles at the temperature of BEC occurrence* is described by equation

$$m_{11} = (h^2 / (2 \cdot \pi \cdot k))^{3/5} \cdot (\rho / 2.612)^{2/5} \cdot T^{3/5}, \quad (1)$$

where  $\rho$  is density;  $h$ ,  $k$  are Planck and Boltzmann constants, respectively. Equation (1) is derived [23] by equating of the temperature ( $T$ ) and the critical temperature of BEC occurrence  $T_c = (h^2 / (2 \cdot \pi \cdot m \cdot k)) \cdot (n / 2.612)^{2/3}$ , where  $m$  and  $n$  are molecular weight and concentration, respectively, of particles of the system (gas) [1, p. 30].

*The mass of particles in a quantum gas — condensate equilibrium can be determined according to the following equation*

$$m = (C_p/C_v) \cdot k \cdot T / C_s^2, \quad (2)$$

where  $C_s$  is a speed of sound in liquid;  $C_p/C_v$  — adiabatic component;  $C_p$  and  $C_v$  — isobaric and isochoric heat capacities of quantum gas. Equation (2) is derived based on the basis of the equilibrium condition  $E_c = E_{i.g.}$ , where  $E_c = m \cdot C_s^2$  is the energy of configuration fluctuations of particles of a liquid participating in quantum gas — condensate reversible transitions;  $E_{i.g.} = (C_p/C_v) k \cdot T$  — fluctuation energy of equilibrium quantum gas particles [23]. There are four levels of energy in equation (2) used for differentiation of the mass of particles ( $m$ ) of a liquid (and for notation index) [23]. Certain adiabatic component of equilibrium quantum gas corresponds to each level of energy:

$(C_p/C_v)_1 = 1$  is the level (1) of quantum gas with critical adiabat;  $(C_p/C_v)_2 = 3/2$  and  $(C_p/C_v)_3 = (5/3)$  — levels of saturated (2) and fully unsaturated (3) ideal monatomic quantum gases by Einstein [13];  $(C_p/C_v)_4 = 2 \cdot \pi$  — the level (4) of quantum gas with thermal wavelength of particles.

The use of speed of sound in equation (2) does not contradict with the BEC — quantum gas equilibrium concept: the speed of sound is used for comparison of Bose-gas and Bose-liquid properties. It is also used in the theory of helium superfluidity [18].

The particle mass values for the BEC — quantum gas equilibrium will be defined as geometric mean of masses determined from equations (1) and (2). Only Einstein saturated ideal quantum gas at BEC equilibrium (2) and quantum gas formed by particles with thermal wavelength (4) will be considered. The case (4) is used to study ultracold gases [8; 16–17; 24], as well as to derive equation for the critical temperature of BEC occurrence [3, p. 614], and for criterion of the ideal gas degeneration [3, p. 608]. The condition of the Einstein saturated ideal quantum gas (2) can not be reached in usual gases [13]. However, in the studied case the condensed condition of substance on a liquid saturation line is considered. Such a condition corresponds to saturated rather than unsaturated quantum gas.

The mass of particles corresponding to the reversible transitions of the BEC — equilibrium of saturated monatomic ideal quantum gas by Einstein is

$$m_{211} = (m_2 \cdot m_{11})^{1/2}. \quad (3)$$

The mass of particles participating in the BEC — quantum gas equilibrium at a level of energy of quantum gas with a thermal wavelength of particles, is calculated by the equation

$$m_{411} = (m_4 \cdot m_{11})^{1/2}. \quad (4)$$

### The mass-spectra comparison

The significance of  $m_{11}$ ,  $m_2$ ,  $m_4$ ,  $m_{211}$  and  $m_{411}$  for H<sub>2</sub>O liquid saturation (obtained by equations (1)–(4) using data from NIST Standard Reference Database [21]) and [9; 25–26], is represented in Fig. 1. Figure 1 shows dependency of  $m$  (atomic mass units) on the temperature ( $T$ , K). The temperature interval is chosen from the supercooled (metastable) liquid state up to the critical temperature. The calculated mass-spectra results are given in comparison with the mass of proton ( $1m_p$ ), the proton pair ( $2m_p$ ) and the 3-proton Efimov resonance ( $3m_p$ ). The proton mass value (1.00739) corresponds to the natural isotopic composition of hydrogen. The values of configuration heat capacity of identical particles ( $C_{c2}$ , J/(mol·K)) are also provided in fig. 1 from the previous work [22]. The values of  $C_{c2}$  were calculated by the equation  $C_{c2} = C_v - C_{vib} - C_{c1} - C_{inf}$ , where  $C_v$ ,  $C_{vib}$ ,  $C_{c1}$ ,  $C_{inf}$  — isochoric, vibrational, configuration distinct particles and infinite heat capacity of H<sub>2</sub>O liquid saturation respectively [21; 22].

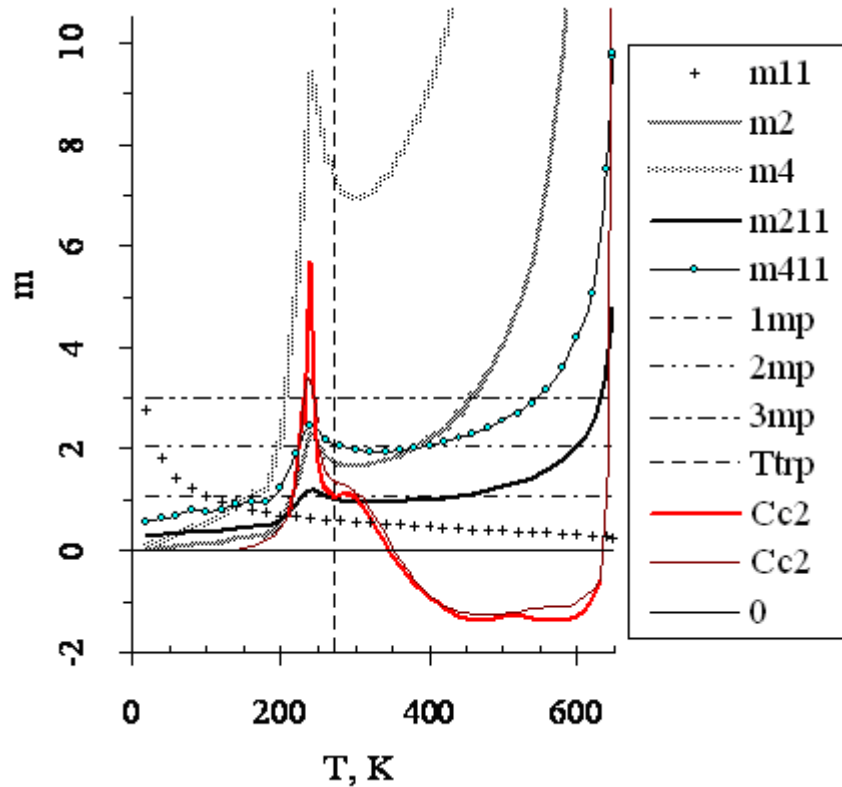


Fig. 1. Dependence on temperature (T, K) found by the equations (1) — (4) modeling masses (m) of particles of H<sub>2</sub>O liquid saturation for the energy levels of Einstein saturated quantum gas (m<sub>2</sub>) and thermal wavelength of particles (m<sub>4</sub>), at temperature of BEC occurrence (m<sub>11</sub>) and BEC — quantum gas equilibrium (m<sub>211</sub>, m<sub>411</sub>): (C<sub>c2</sub> — configuration heat capacity of identical particles (J/ (mol·K)) by data [22]; T<sub>tp</sub> — triple point temperature)

The values of m<sub>11</sub> decrease smoothly while the temperature increases, which can be noticed within m<sub>411</sub>(T) and m<sub>211</sub>(T) functions. These functions depend on the temperature less than m<sub>4</sub> and m<sub>2</sub> do. In a temperature course of functions m<sub>411</sub>(T) and m<sub>211</sub>(T) the constancy of the mass ratio  $m_{411} / m_{211} = (m_4 / m_2)^{0.5} = (4\pi / 3)^{0.5} \approx 2.04665$  is also shown. In a wide temperature range the values of m<sub>211</sub> are nearly equal to the proton mass (1m<sub>p</sub>), and values of m<sub>411</sub> are nearly equal to the mass of proton pair (2m<sub>p</sub>) and m<sub>2</sub>. The temperature area, in which the values of m<sub>411</sub> and m<sub>2</sub> are nearly equal to the proton pair mass, is more extensive in the m<sub>411</sub>(T) case than in the m<sub>2</sub>(T) case. At temperatures 239–240K of m<sub>211</sub>(T) and m<sub>411</sub>(T) functions have maximum. In this temperature range the configuration heat capacity of identical particles (C<sub>c2</sub>(T), see fig. 1) has a sharp maximum. In the field of temperatures 350-630K the C<sub>c2</sub>(T) function becomes negative. In this temperature range m<sub>411</sub> and m<sub>211</sub> increase with the temperature. The greatest masses of m<sub>411</sub> and m<sub>211</sub> correspond to the critical temperature.

### Mass-spectra and configuration capacity of identical particles

Features of functions m<sub>211</sub> (T) and m<sub>411</sub> (T) are used for interpretation of extreme in temperature dependence of a configuration heat capacity of identical particles of H<sub>2</sub>O liquid saturation C<sub>c2</sub>(T).

### The temperature range of 273-300 K

There is a minimum of  $C_{c2}(T)$  function at the temperature of triple point of H<sub>2</sub>O water (see fig. 1). There is a maximum of  $C_{c2}(T)$  at 285K. Both extremes are poorly expressed and their presence is challenged because  $C_{c2}$  values are derived based on a difference of large numbers. However, at an example when the pressure is 100 MPa and the temperature is 285 K there is a maximum in a temperature dependence of isochoric heat capacity  $C_v(T)$  [21]. The  $C_{c2}(T)$  function has an extreme on a line of a liquid saturation provides that the function  $C_{c2}(T)$  is more sensitive to changes of liquid properties, rather than an isochoric heat capacity temperature function  $C_v(T)$ . This, therefore, can be the basis to consider H<sub>2</sub>O extremes for a discussion.

The temperature of  $C_{c2}(T)$  maximum (285 K) practically coincides with the temperature (286 K), at which the value of  $m_{411}$  equals to the mass of proton pair. The mass of proton pair can be derived using a modeling system containing one cluster (2H). It can also be derived based on the system containing two clusters (4H; 4H). It means, that the  $C_{c2}(T)$  maximum corresponds to modeling systems containing bosons. Transitions from even-even system (4H; 4H) to the nearest even-odd systems ((3H; 4H) or (4H; 5H)) result in an increase of the fermion content and in a decrease of configuration heat capacity of  $C_{c2}$  identical particles. Thus, the  $C_{c2}(T)$  maximum at the temperature of 285 K can be classified as a boson peak. The obtained result within the framework of the BEC-quantum gas equilibrium concept coincides with those, obtained on the basis of the analysis of frequencies of configuration vibrations in equilibrium of a quantum gas — condensate [22].

At the temperature of 273.16 K, corresponding to the  $C_{c2}(T)$  minimum, the  $m_{211}$  value is greater than the mass of proton. According to the BEC-quantum gas equilibrium concept it is possible to explain the deviation of  $m_{211}$  from the proton mass at the temperature of the  $C_{c2}$  minimum. In considered temperature range  $m_{211}(T)$  rises with the temperature decrease. The proton mass can be obtained based on (1H) and (2H; 2H) modeling systems. The last system (2H; 2H) is classified as even-even modeling system, which contains bosons. The presence of bosons causes the values of configuration heat capacity  $C_{c2}$  to be positive (increased comparatively to the values at 273.16 K). The decrease of the heat capacity  $C_{c2}$ , observed at temperatures lower than 275 K is related to an increase of the fermion content in the modeling system. Indeed, it is possible to obtain the mass exceeding the proton mass in an even-odd modeling system (2H; 3H). In such system the fermion content is higher, than in (2H; 2H) system. The mass close to  $m_{211} = 1,01425$  corresponding to the  $C_{c2}(T)$  minimum can be obtained from various modeling systems. This characterizes such modeling systems at 273.16K as variable. For example, modeling systems (2H; 3H; 1HO; 1H<sub>4</sub>O; 1H<sub>4</sub>O), (2H; 3H; 1M; 1M; 2M; 3OH), and (2H; 3H; 1M; 1M; 2OH; 3M), where  $M \equiv H_2O$ , correspond to masses 1.0143, 1.0146, and 1.0141. The variability testifies high probability of modeling system realization.

### The temperature range of 300–400 K

The mass of  $m_{411} = 1,9459$ , corresponding to the  $m_{411}(T)$  minimum at 329K, can be obtained as given in the model system, in which the least cluster contains two or greater number of protons, which is in agreement with proton pairs in a H<sub>2</sub>O liquid found earlier [22]. In the range of 329K the  $C_{c2}$  heat capacity is positive, therefore proton pairs could be used in the modeling of the minimal value (0,9508) of the  $m_{211}(T)$  function. For example, the even-even (2H; 2H; 1OH) model system used in calculation of the mass of the proton H<sup>+</sup> and OH<sup>-</sup> ion gives a value of 0,951, which is close to the desired value.

When the temperature rises above 329K the mass of particles  $m_{211}$  and  $m_{411}$  increases and even-even system are giving way to even-odd model systems. This leads to an increase of fermion clusters contribution to configurational heat capacity of identical particles  $C_{c2}$  as well as its transition to negative values. Fermions, by definition, [6], interfere with a negative sign, which manifests itself in the configuration of the heat capacity of identical particles  $C_{c2}(T)$ . The role of all clusters of the model system appears in the process of the  $C_{c2}$  decrease. For example, at 350K the  $m_{411} = 1,957$  value is modeled by the system (3H; 1O; 1HO; 1HO), which contains one bosonic cluster (1O) formed by  $O^{2-}$  particles among three fermion clusters. At higher temperatures (361-362K) the  $m_{411} = 1,971$  value is modeled by the fermionic system (3H; 1HO; 1HO; 1HO).

### The temperature range of 200–273 K

High temperature (right) wing of the  $C_{c2}(T)$  maximum is observed at temperatures from 239–240 K to 273,16 K. The mass of 1,2088698 corresponds to the model system (2H; 3H). This mass exceeds the maximum value of  $m_{211}$  (1,19272), which corresponds to 239 K. In this regard, the  $m_{211}$  values of all of the temperatures of the right wing of the  $C_{c2}(T)$  maximum can be calculated as at the temperature of 273,16 K based on the even-odd system (2H; 3H). For example the mass of 1,1929, close to the value of  $m_{211}$  at the maximum  $C_{c2}(T)$ , corresponds to the system (2H; 3H; 5M). Even-odd (2H; 3H) systems are also suitable for describing the temperature dependence of the  $m_{211}$  in the left (low temperature) wing of the  $C_{c2}(T)$  maximum. However, (2H; 3H) systems can not explain the reasons for sharp  $C_{c2}(T)$  rise as it approaches the maximum in the configurational heat capacity of identical particles. The problem can be solved by increasing hydrogen parity in the process of changing the composition of model systems. The even-odd system (2H; 3H) has to be consistently replaced with (2H; 4H) and (2H; 6H) even-even model systems as the temperature approaches the  $C_{c2}(T)$  maximum. At the  $C_{c2}(T)$  maximum the following systems can be realized (2H; 4H; 1O; 2O) and (2H; 6H; 1OH; 1OH; 1OH), which correspond to 1,1930 and 1,1931 values. In the case of ethane the (2H; 4H) model was attributed to the number of critical systems [23]. In this case the (2H; 4H) system is also critical, since the next even-even (2H; 6H) system corresponds to an equivalent (3H; 3H; 1OH; 1OH; 1OH) odd-odd model in which all clusters are fermionic.

At large values of the inductive capacity of water, and at differences in the electronegativity of hydrogen and oxygen, all the clusters (3H; 3H; 1OH; 1OH; 1OH) in odd-odd model system are electrically charged particles ( $(3H)^{3+}$ , OH<sup>-</sup>) and, accordingly, are fermions. *The possibility of transition of model system containing predominantly boson clusters to the system of a fermionic structure is characteristic to liquid H<sub>2</sub>O.* Further, it is one of the reasons for the formation of *spiked maximums* of configurational heat capacity of identical particles  $C_{c2}(T)$  at 239–240 K. *Boson-fermion contrasts* reflect, for example, (2H; 4H; 1O; 2O) and (3H; 3H; 1OH; 1OH; 1OH) systems, which simulate the  $m_{211}$ -mass values within  $C_{c2}(T)$  peak at 239–240 K temperatures.

### The temperature range of 400–620 K

The shape of an extensive  $C_{c2}(T)$  minimum can be explained by the equilibrium BEC-quantum gas concept in the temperature range of 400–620 K. From Fig. 1 it follows that the trend line of  $C_{c2}(T)$  function has a wave-like shape in these three segments of temperatures. A weak maximum, corresponding to 510–520 K temperatures, is surrounded by two nearly symmetrically located minima. The first of these is located at 460–470 K, and the second is in the temperature range of 560–570 K.

The  $m_{211}(T)$  function at temperatures from 402 K to 484 K has the same values as at 275 K to 239–240K temperatures. Therefore, the  $m_{211}(T)$  are modeled by (2H; 3H) even-odd systems. (2H; 3H; 1OH) and (2H; 3H; 1M) model systems, for example, correspond to  $m_{211}(T)$  values in the temperature range of 460–470 K. In the temperature range of 484 K the  $C_{c2}(T)$  function increases with higher temperatures. In this case, the mass  $m_{211} = 1,1927$  is simulated by model system with two equivalent forms (3H; 3H; 1OH; 1OH; 1OH) and (2H; 6H; 1OH; 1OH; 1OH). All the components of one system are classified as fermions. They are classified as bosons (2H; 6H) in another system. In the temperature range of 470–520 K the  $m_{211}$  mass can also be obtained by other containing bosons model systems. The implementation of (2H; 4H) even-even systems such as (2H; 4H; 1HO); (2H; 4H; 1M), (2H; 4H; 2O) and alike, contributes to the maximum of  $C_{c2}(T)$  in the temperature range of 510–520 K. A significant contribution to the formation of the  $C_{c2}(T)$  maximum is also made by (4H) systems. With further increase in temperature the contribution of even-odd model systems to  $m_{211}$  and  $m_{411}$  masses increases, which leads to some decrease in  $C_{c2}$  and the observation of a minimum of  $C_{c2}(T)$  at 560–570 K. Such factors as high variability of model systems, the possibility of smooth and continuous temperature change in the mass of particles in mass spectra of  $m_{211}(T)$  and  $m_{411}(T)$ , and dissociation of molecules into ions allow *smoothing the boson-fermion variation of  $C_{c2}(T)$  on the H<sub>2</sub>O liquid saturation line in the temperature range of 400–620 K.*

### The near-critical temperature region

At the critical temperature ( $\approx 647,27\text{K}$ ) the highest value  $m_{411} \approx 9,8$  is achieved. In order to model this number by using hydrogen clusters, they must contain at least 10 protons. In such model system (10H; 20M), along with 10-atomic resonances (10H), it is necessary to use the 20-molecule (20M) Efimov resonance. To reduce the size of the second cluster it is necessary to increase the mass of the first cluster. The  $m_{411} = 9,8$  value can be modeled by, for example, (12H; 3HO) system, in which the 12H cluster mass commensurate with  $2/3$  of the mass of water molecules. If using only H<sub>2</sub>O molecules in the cluster, the model system will be (1M; 2M; 3M). It is known that the mass of the liquid particles increases at the critical temperature. However, in this case, the (3M) cluster is a classic three-body Efimov resonance, when the (2M) cluster has the resonance nature, formed by the coherent motion of particles. Bosonic nature of the molecules characterizes high meanings of configurational heat capacity as the boson peak. However, the configurational heat capacity of H<sub>2</sub>O is less than this of D<sub>2</sub>O [22]. This fact was attributed to the manifestation of fermions in the configurational heat capacity of identical particles of H<sub>2</sub>O. Indeed, for example, at the critical temperature, the  $m_{211}$  (4,78) value can be simulated by systems containing, in addition to bosons, fermion clusters: (5H; 6O) = (10H; 10H; 6O), (1M; 1M; 1M; 3O; 3O), (1HO; 1M; 1M; 3HO; 3HO). The possibility of representing  $m_{411}$  and  $m_{211}$  masses in the form of models of different composition characterizes model systems at critical temperature as a variable. *The transformation of proton resonances to molecular resonances is one of the factors which reduces the scale of BEC manifestations in liquid H<sub>2</sub>O*, as opposed to, for example, cold gases [24].

### Mass spectra and the structure

Features of  $m_{211}(T)$  and  $m_{411}(T)$  mass spectra are used for quantitative estimation of H<sub>2</sub>O liquid structure on the saturation line. Mass spectra contain resonances that are formed due to the coherent motion of identical particles of liquid. The de Broglie waves, related to the BEC-

quantum gas equilibrium, propagate with the speed of sound ( $C_s$ ). In this case, the de Broglie wavelength ( $\lambda$ ) is comparable to the intermolecular and interatomic distances ( $d$ ) of the liquid. The phenomenon of wave diffraction and interference can be used to study the liquid structure. To realize this possibility the particle mass ( $m$ ) of de Broglie wave sources and receivers will be compared with  $m_{211}$  and  $m_{411}$  masses.

To determine the mass of particles ( $m$ ) the de Broglie wavelength  $\lambda = h / (m \cdot C_s)$  is used in Bragg's law Equation  $2 \cdot d \cdot \sin(\alpha) = n \cdot \lambda$ , where  $\alpha$  is a slip angle, and  $n$  is the order of diffraction. In this case, the mass is dependent on the angle  $\alpha$ :

$$m = n \cdot h / (2 d C_s \sin(\alpha)). \quad (5)$$

Diffraction waves interfere. In the calculations of fluctuation intensity ( $J, J_1, J_2$ ) in accordance with the interference:  $J = J_1 + J_2 + 2 \cdot (J_1 \cdot J_2)^{1/2} \cdot \cos(\Phi)$ , where  $\Phi$  — the difference of phase fluctuations, an additional angle ( $\Phi$ ) has to be considered. To account for possible angle variations ( $\alpha$  and  $\Phi$ ) the structural features of liquids will be taken into account. In these liquids, unlike solids, the short-range order takes place, which undergoes temperature changes. In this regard, «the composition» and structure of the first coordination sphere of the molecules we mainly taken into account.

The molecules of the first coordination sphere of water  $H_2O$  are divided [14, 20] into fractions:  $f_1$  and  $f_2$  — for intermolecular distances 0,28 and 0,33 nm. Multiplication of fractions  $f_1$  and  $f_2$  on the coordination number  $z$  gives the number of molecules of the first ( $n_1$ ) and the second ( $n_2$ ) type in the first coordination sphere of water  $z = n_1 + n_2$ .

It was found [5] that in a wide temperature range  $n_1$  remains constant and equal to the value (2,36), the reverse value is equal to the percolation threshold for the tetrahedral lattice sites (0,43 [2, p. 436]). Since, by definition, the percolation threshold is  $n_1 = z_1 - 1$ , where  $z_1$  — a coordination number of molecules in lattice sites, then  $z_1 = n_1 + 1$ . By analogy,  $z_2 = n_2 + 1$  is the coordination number of molecules in position 2 (in the «interstices»). The number  $z$  is the vector sum of  $z_1$  and  $z_2$  [5], in contrast to the algebraic sum of  $z = n_1 + n_2$ . Thus,  $z, z_1,$  and  $z_2$  form a vector triangle. Parameters of the triangle associated with the composition and structure of the first coordination sphere.

The vector triangle parameters will be used to describe the phenomena of diffraction and interference of de Broglie waves. Let the straight line to coincide with the interatomic or intermolecular distance  $d$  at the right angle of one side of the triangle. The intensity will be represented in mass units as the product of mass  $m$  in (5) and fractions. The indexes (0, 1, 2) will be used to describe fractions ( $f_0 = f_1 + f_2; f_1; f_2$ ) and angles ( $F_0; F_1; F_2$ ) at the opposite sides of  $z, z_1$  and  $z_2$ . Indexes will be assigned to letters ( $i = 0; j = 1, k = 2$ ). Angles ( $\alpha$  and  $\Phi$ ) will be equated to corners of the vector triangle. The chosen succession let us to account for all the angles of the vector triangle in each computation of the mass of particles involved in the phenomena of diffraction and interference of de Broglie waves.

The result of interference described by equation (taking into account the summation of mass ( $m$ ) to (5) and their reciprocals ( $m^{-1}$ ):

$$m_{ijk} = ((m_i)^b \cdot f_j + (m_j)^b \cdot f_i + (-1)^c \cdot 2 \cdot ((m_i)^b \cdot f_j \cdot (m_j)^b \cdot f_i)^{1/2} \cdot \cos(F_k))^b, \quad (6)$$

where  $b$  can be +1 and -1;  $c$  — may be 1 and 2, the first and second indexes ( $m_{ijk}$ ) — characterize the slip angle and the fraction in the first term, as well as the fraction and the slip angle in the second term. The third index characterizes phase (in the third term) of the right-hand side of the equation (6). After index permutation the first index transfers to the place of the third one. As a



result, equation (6) gives 12 meanings of the mass of particles, which are associated with de Broglie waves involved in the phenomena of diffraction and interference.

Comparing computational results of the equation (6) with the results of equations (1) – (4) the distance  $d$  can be estimated.

### Mass spectra and intermolecular distances

In this paper, based on the results obtained from (6) a geometric mean ( $m_{g.m.}$ ) of mass was found when  $b = +1$ ,  $b = -1$  and  $b = (+1 \text{ and } -1)$  with a parameter  $c = 1$ ;  $c = 2$ ;  $c = (1 \text{ and } 2)$ . Such an averaging gives 15 masses ( $m_{g.m.}$ ) of particles. Configuration fluctuations of the fluid particles reduce the difference between fractions  $f_1$  and  $f_2$ . In this paper  $f_1 = f_2 = 0,5$  was accepted. Computational results of  $m_{g.m.}$  involved in the diffraction and interference, was compared with the  $m_{211}$  mass. Using this method, the distance  $d$  (nm) for H<sub>2</sub>O on the liquid saturation line was determined, which is shown in Fig. 2 as  $d(T)$  functions (lines 1–15). The effective radius of the molecules ( $r$ ) and various data in the literature [4, 14, 20, 25, 26] are also presented.

From Fig. 2 it follows that most of the calculated  $d(T)$  functions (lines 1–15) are convex to the temperature axis (T, K).

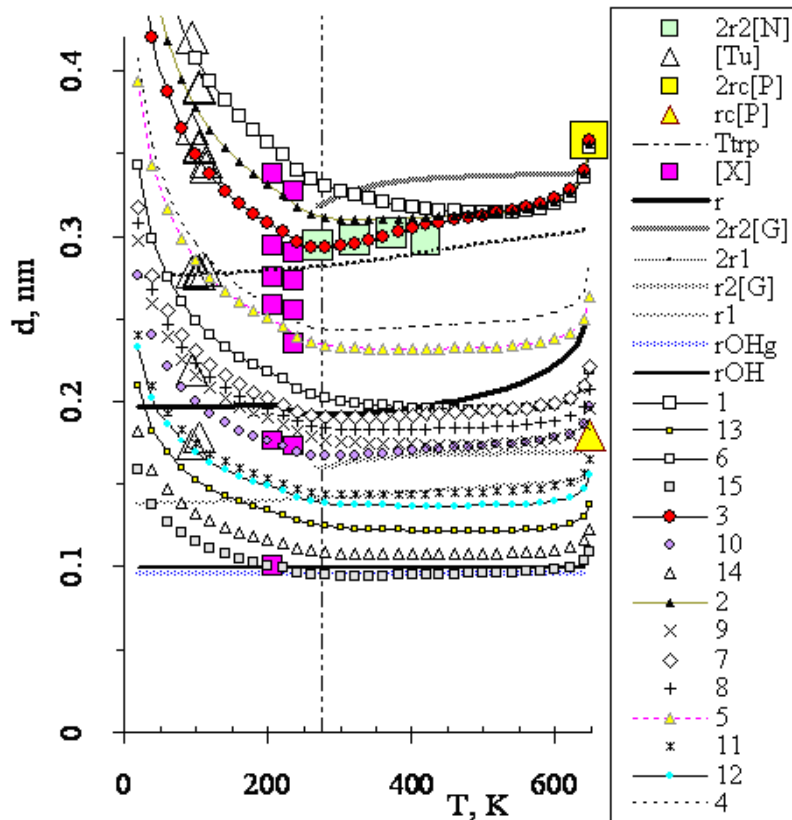


Fig. 2. Intermolecular distances ( $d$ , nm) of H<sub>2</sub>O on the saturation line of the liquid at temperatures (T, K) according to equation (6) calculations, when  $f_1 = f_2 = 0.5$  and  $m_{g.m.} = m_{211}$ , (lines 1–15) and literature data: ([G; N; P; Tu; X] — in [14, 20, 4, 25, 26];  $r_1, r_2, r_c, 2r_1, 2r_2, 2r_c$  — radius and intermolecular distances of the first and second types of molecules and critical fluid;  $r_{OH}, r_{OHg}$  — the distance between the atoms of OH-liquid and H<sub>2</sub>O-gas;  $r$  — effective radius of the liquid molecules;  $T_{trp}$  — triple-point temperature)

The lowest meanings of  $d(T)$  (lines 14–15) are in the region of interatomic distances of liquid ( $r_{OH}$ ) and gas ( $r_{OHg}$ ). The greatest meanings of  $d(T)$  (lines 1–3) are located in the intermolecular distances of the first ( $2r_1$ ) and second ( $2r_2$ ) types of molecules. Lines 10–11 practically possess the same features of the temperature dependence of the molecular radius ( $r_2$ ) and ( $r_1$ ). An agreement of obtained  $d(T)$  distances was found for stable, supercooled and critical liquids.

The angular characteristics ( $F(T)$ ) of molecules and their relative position in the liquid can be estimated based on  $d(T)$  of, for example, adjacent lines. From these estimations it follows, that the angle of  $H_2O$  gaseous molecules ( $104,52^\circ$ ) is the limit of the  $F(T)$  function at the critical temperature. While, characteristic angles of tetrahedral ( $109,47^\circ$ ), hexagonal ( $120^\circ$ ), as well as the adjacent angle of pentagonal ( $108^\circ$ ) coordination appear as the  $F(T)$  function limit at low temperatures only.

From the analysis of the temperature dependence of the configurational heat capacity of identical particles  $C_{c2}(T)$  and intermolecular distances  $d(T)$ , calculated on the basis of our proposed concept of equilibrium BEC-quantum gas, it follows that within the properties and structure of  $H_2O$  on the saturation line of liquid the Bose-Einstein condensate is manifested up to the critical temperature. In addition, such a manifestation is confirmed by the expression of classical (3-body) Efimov resonances and by resonances that contain a large number of particles (protons,  $H_2O$  molecules).

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