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Clusterization of water molecules as deduced from statistical mechanical approach*

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Abstract: Using the methods of statistical mechanics we have shown that a homogeneous water network is unstable and spontaneously disintegrates to the nonhomogeneous state (i.e. peculiar clusters), which can be treated as an ordinary state of liquid water. The major peculiarity of the concept is that it separates the paired potential into two independent components - the attractive potential and the repulsive one, which in turn should feature a very different dependence on the distance from the particle (a water molecule in the present case). We choose the interaction potential as a combination of the ionic crystal potential and the vibratory potential associated with the elastic properties of the water system as a whole. The number $\aleph$ of water molecules that enters a cluster is calculated as a function of several parameters, such as the dielectric constant, the mass of a water molecule, the distance between nearest molecules, and the vibrations of nearest molecules in their nodes. The number of H_2O molecules that comprise a cluster is estimated as about $\aleph \approx 900$, which agrees with the available experimental data.

Keywords: Water, clusters, statistical mechanics
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1 Introduction

The dielectric response of liquid water in the frequency range from 10 up to 1000 cm^{-1} has shown [1, 2] that the dielectric spectrum of water is characterized by a complex permittivity in the microwave region and two absorption bands in the far infrared region;

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the maxima have been recorded at 200 and 700 cm^{-1}. Generally, two-component models provide a simple way of accounting for many thermodynamic anomalies of liquid water [2]. The major results obtained are the following [2]: the dielectric relaxation is successfully represented by a biexponential model with a fast (< 0.3 ps) and a slow (> 2 ps) decay time; the slow decay time is consistent with structural relaxation of water and the temperature dependence of the slow relaxation time allows modelling from a singular point at 328 K.

These results correlate very well with our study on the two-component water model [3, 4, 5], the existence of a temperature singularity point and the existence of thermodynamic anomalies of water. Over twenty years ago Zelepukhin and Zelepukhin [6, 7] (see also Refs. 3,5) established that the removal of part of the gases dissolved in water under normal temperature and pressure conditions change its biological activity. The measurement [3] of spin-lattice relaxation rates for protons in water assuming that the latter contains both tightly and loosely bound molecules, in fact, confirmed the existence of the critical value T_c. By our measurements and estimates T_c has varied from about 290 K to 338 K, depending on the kind of water specimens (see Refs. 3 to 7).

Very interesting data were obtained by Gordeev and Khaidarov [8]: they detected ostensible density fluctuations, globules to 3 nm in size comprising up to 10^3 molecules, and noted that the bulk water could be regarded as a polycrystal-ferroelectric with a domain structure in time ~ 100 ps. Similar results were presented by Luck [9] (for other references see Ref. [5]).

In paper [10] we proposed an approach that allowed the investigation of cluster formation in various systems of interacting particles; namely, we developed rather a very general method of statistical behavior of particles that obeyed Fermi or Bose statistics and whose interaction could be subdivided into two opposite kinds of interaction, i.e. attractive and repulsive (those were electrons on the surface of a liquid helium film, charged particles in Yukawa's potential and stars that gravitationally interacted on the background of the expansion of the universe). The method [10] enabled us to reduce the quantity of variables that initially described the Fermi and Bose systems in question and a new canonical variable, which characterized the nascent nonhomogeneous state (i.e. clusters), automatically arose as a logical consequence of the particles' behavior. Then the methodology was successfully employed [5] for the description of systems that could be characterized by classical statistics.

In this paper we consider how the methodology [10] can describe clusters in the network of water molecules.

2 Hamiltonian and partition function

Let us assume that the water network is described by a lattice model in which nodes are filled by a water molecule or empty. Therefore, the relevant variable, the filling number n_s, can be equal to 1 (if a water molecule is at the sth state) or equal to 0 (if the sth site is empty). Although such a modelling system rather looks like a cubic one, we

are not attached to a concrete kind of symmetry. The results anticipated will be quite appropriate for a real water network, because at times less than 10^{-11} s the latter in fact can be considered as a typical unregulated crystal lattice, which, at least at the first approximation, allows the description in terms of averaged parameters of the ordered lattice.

Let us represent the Hamiltonian of the water system in the form

$$H\left(n\right)=\sum_{s}E_{s}n_{s}-\tfrac{1}{2}\sum_{s,\,s'}V_{ss'}n_{s}n_{s'}+\tfrac{1}{2}\sum_{s,\,s'}U_{ss'}n_{s}n_{s'} \tag{1}$$

where E_s is the additive part of the particle energy in the sth state. The main point of our approach is the initial separation of the total atomic/molecular potential into two terms: the repulsive and attractive components. So, in the Hamiltonian (1) $V_{ss'}$ and $U_{ss'}$ are respectively the paired energies of attraction and repulsion between particles located in the states s and s'. It should be noted that the signs before the potentials in expression (1) specify the proper signs of the attractive and repulsive paired energies, which means that both functions $V_{ss'}$ and $U_{ss'}$ in expression (1) are positive. The partition function of the system

$$Z=\sum_{\{n\}}\exp\left(-H\left(n\right)/k_{\mathrm{B}}T\right) \tag{2}$$

may be presented in the field form

$$\begin{aligned}Z=&\int_{-\infty}^{\infty}D\phi\int_{-\infty}^{\infty}D\psi\sum_{\{n\}}\exp\Big[\sum_{s}\left(\psi_{s}+i\phi_{s}\right)n_{s}\\&-\sum_{s}\tilde{E}_{s}n_{s}-\frac{1}{2}\sum_{s,\,s'}\left(\tilde{U}_{ss'}^{-1}\phi_{s}\phi_{s'}+\tilde{V}_{ss'}^{-1}\psi_{s}\psi_{s'}\right)\Big]\end{aligned} \tag{3}$$

due to the following representation known from the theory of Gauss integrals

$$\exp\left(\frac{\rho^{2}}{2}\sum_{s,\,s'}W_{ss'}n_{s}n_{s'}\right)=\mathrm{Re}\int_{-\infty}^{\infty}D\vartheta\,\exp\left[\rho\sum_{s}n_{s}\vartheta_{s}-\tfrac{1}{2}\sum_{s,\,s'}W_{ss'}^{-1}\vartheta_{s}\vartheta_{s'}\right] \tag{4}$$

where $D\,\vartheta\equiv\prod_{s}\sqrt{\det||W_{ss'}||}\sqrt{2\pi}\,d\vartheta_{s}$ implies the functional integration with respect to the field ϑ; $\rho^{2}=\pm1$ in relation to the sign of interaction ($+1$ for attraction and -1 for repulsion). The dimensionless energy parameters $\tilde{V}_{ss'}=V_{ss'}/k_{\mathrm{B}}T$, $\tilde{U}_{ss'}=U_{ss'}/k_{\mathrm{B}}T$, and $\tilde{E}_{s}/k_{\mathrm{B}}T$ are introduced into expression (3). Further, we will use the known formula

$$\frac{1}{2\pi i}\oint dz\,z^{\,N-1-\sum_{s}n_{s}}=1, \tag{5}$$

which makes it possible to fix the quantity of particles in the system, $\sum_{s}n_{s}=N$, and, consequently, we can pass to the consideration of the canonical ensemble of N particles. Thus the partition function (3) is replaced for

$$\begin{aligned}Z=\mathrm{Re}\,\tfrac{1}{2\pi i}\oint dz\int D\phi\int D\psi\exp\{-\tfrac{1}{2}\sum_{s,\,s'}\left(\tilde{U}_{ss'}^{-1}\phi_{s}\phi_{s'}+\tilde{V}_{ss'}^{-1}\psi_{s}\psi_{s'}\right)\\+\left(N-1\right)\,\ln z\}\times\sum_{\{n_{s}\}=0}^{1}\exp\{\sum_{s}n_{s}\left(\psi_{s}+i\,\phi_{s}-\tilde{E}_{s}\right)-\ln z\}.\end{aligned} \tag{6}$$

Summing over n_s we obtain

$$Z = \mathrm{Re}\frac{1}{2\pi i}\int D\phi \int D\psi \oint dz\, e^{\,S(\phi,\psi,\,z)} \tag{7}$$

where

$$\begin{aligned} S = \sum_s \Bigg\{ & -\tfrac{1}{2}\sum_{s'}\left(\tilde{U}^{-1}_{ss'}\,\phi_s\phi_{s'} + \tilde{V}^{-1}_{ss'}\,\psi_s\psi_{s'}\right) \\ & +\eta\,\ln\left|1+\tfrac{\eta}{z}\,e^{-\tilde{E}_s}e^{\psi_s}\cos\phi_s\right|\Bigg\} + (N-1)\;\ln z. \end{aligned} \tag{8}$$

Here, the symbol η characterizes the kind of statistics: Bose ($\eta = +1$) or Fermi ($\eta = -1$). Let us set $z = \xi + i\zeta$ and consider the action S on a transit path that passes through the saddle-point at a fixed imaginary variable $\mathrm{Im}\, z = \zeta_0$. In this case S may be regarded as the functional that depends on the two field variables ϕ and ψ, and the fugacity $\xi = e^{-\mu/k_{\mathrm{B}}T}$ where μ is the chemical potential.

In a classical system the mean filling number of the sth energy level obeys the inequality

$$n_s = \frac{1}{\xi}\,e^{-\tilde{E}_s} = e^{(\mu - E_s)/k_{\mathrm{B}}T} \ll 1 \tag{9}$$

(note the chemical potential $\mu < 0$ and $|\mu|/k_{\mathrm{B}}T \gg 1$). By this means, we can simplify expression (8) expanding the logarithm into a Taylor series in respect to the small second member. As a result, we get the action that describes the ensemble of interacting particles that obey Boltzmann statistics

$$S \cong \sum_s \left\{ -\tfrac{1}{2}\sum_{s'}\left(\tilde{U}^{-1}_{ss'}\,\phi_s\phi_{s'} + \tilde{V}^{-1}_{ss'}\,\psi_s\psi_{s'}\right) + \frac{1}{\xi}\,e^{-\tilde{E}_s}e^{\psi_s}\cos\phi_s \right\} + (N-1)\ln\xi. \tag{10}$$

The extremum of the functional (10) should be realized at solutions of the equations $\delta S/\delta\phi_s = 0$, $\delta S/\delta\phi_s = 0$ and $\delta S/\delta\xi = 0$. The corresponding equations appear as follows

$$\sum_{s'}\tilde{U}^{-1}_{s'}\phi_{s'} = -\frac{2}{\xi}\,e^{-\tilde{E}_s}e^{\psi_s}\sin\phi_s, \tag{11}$$

$$\sum_{s'}\tilde{V}^{-1}_{s'}\psi_{s'} = -\frac{2}{\xi}\,e^{-\tilde{E}_s}e^{\psi_s}\cos\phi_s, \tag{12}$$

$$\frac{1}{\xi}\sum_{s'}\,e^{-\tilde{E}_{s'}}e^{\,\psi_{s'}}\cos\phi_{s'} = N-1. \tag{13}$$

We introduce the designation

$$\aleph_s = \frac{1}{\xi}\,e^{-\tilde{E}_s}e^{\psi_s}\cos\phi_s, \tag{14}$$

which in fact is a "combined variable", because it is includes the field variables ψ_s and ϕ_s and the fugacity ξ. Inserting $\aleph_s$ into the left hand side of equation (13) we obtain

$$\sum_s \aleph_s = N-1. \tag{15}$$

In other words, $\aleph_s$ indicates the number of water molecules in the sth state and, therefore, the variable $\aleph_s$ represents the number of molecules in the sth cluster.

Variables $\aleph_s$ and ξ allow rewriting the action (10) as follows (see technical details in Ref. [10])

$$S = -\,2\sum_{s,\,s'}\left[\tilde{V}_{s\,s'}\,\aleph_s\aleph_{s'} + \tilde{U}_{s\,s'}\,\aleph_s\aleph_{s'}\left(\frac{e^{-2\tilde{E}_s+4\sum_{s'}\tilde{V}_{s\,s'}\,\aleph_{s'}}}{\xi^2\,\aleph_s^2} - 1\right)\right] + \sum_s \aleph_s\,(1+\ln\xi). \tag{16}$$

Nevertheless, if we introduce a combined variable designated as $\aleph_s = \aleph$ for each of the clusters, we can then pass to the continuum presentation of the action (16), which results in the action (see Ref. [10] for details)

$$S = K \times \{\,2\aleph\,(a-b) - 2\frac{1}{\xi^2}\,a\,e^{-2\tilde{E}+4b\aleph} + \aleph\ln\xi\,\} \tag{17}$$

where K is the number of clusters in the system studied. Here the functions a and b are given by

$$a = 3\int_1^{\aleph^{1/3}}\tilde{U}\,(gx)\;x^2dx, \qquad\qquad b = 3\int_1^{\aleph^{1/3}}\tilde{V}\,(gx)\;x^2dx \tag{18}$$

where the potentials $\tilde{U}$ and $\tilde{V}$ depend on the dimensionless variable $x = r/g$ and g is the distance between water molecules. All of the prior consideration is correct in the approximation $\aleph \gg 1$.

The second term in the right-hand side of expression (17) is the smallest one, because $e^{-2\tilde{E}}/\xi^2 \ll 1$, and is omitted hereinafter. Thus we shall start from the action

$$S = K \times \{2\,\aleph\;(a-b) + \aleph\;\ln\xi\}. \tag{19}$$

The extremum (minimum) of the action (19) is reached at the meaning of $\aleph$, which is found from the equation $\delta S/\delta\aleph = 0$ and satisfies the inequality $\delta^2 S/\delta\aleph^2 > 0$. The value $\aleph = \aleph_0$ obtained in such a way will correspond to the number of particles that form a cluster.

3 Realistic potentials

Since we wish to study the interaction of molecules in water, we shall write a model intermolecular potential. The infrared spectrum of water shows strong activity in the range of the hydrogen bond O−H$\cdots$H, i.e. around 200 cm^{-1}, which means that water molecules are characterized by a strong ionic polarizability. This automatically signifies that the study of the statistical behavior of a water systems around 200 cm^{-1} (e.g. in the range of frequencies $\nu \approx 1$ to 400 cm^{-1}) might be carried out in the framework of

a typical ionic lattice. Therefore this allows us to simulate the pair potential of a water molecule in the form of an ionic crystal potential

$$W_{\mathrm{H_2O-H_2O}} = V_0 \ \exp\left(-\, r/g\right) - \alpha \frac{e^2}{4\pi\varepsilon_0 \varepsilon\, r}. \tag{20}$$

Here V_0 is the energy that describes the short-range repulsion, or paired repulsion of two nearest molecules; g is the radius of this repulsion force, which we below identify with a typical distance between nearest molecules (the lattice constant); α is Madelung's constant that, as a rule, falls within the range from unity to two (see, e.g. Kittel [11]); ε is the dielectric constant (which, however, is absent in the case of ionic crystals); and lastly r is the distance between interacting particles.

In the preliminary study [5] we started just from the potential (20), though at the transition from the discrete to the integral representation some details were not taken into account. The transition to the integral representation was studied with great accuracy in Ref. [10], although for completely different systems. In the present work we shall combine the considerations performed in Refs. [5, 10] and, moreover, the paired potential (20) requires supplementation by one more term. Exactly what kind of a term is needed?

In a series of recent works a submicroscopic concept of physics has been presented, which allows the reconsideration of the foundations of the fundamentals. Namely, a detailed mathematical theory of the real physical space has been constructed by Bounias and the author [12–14], which becomes a remarkable basis for submicroscopic mechanics of particles developed in papers [15–21]. Thus submicroscopic mechanics of particles are developed in the real physical space, which means that such mechanics is complete deterministic and is specified by a short-range action that is ensured by light carriers (or quasi-particles, or elementary excitations) of the space, called "inertons". The theory is in force from sub-Planck distances to cosmic ones. By the theory, any canonical particle is surrounded by its own cloud of inertons, which transfers inert properties of the particle. The cloud spreads to a distance

$$\Lambda = \lambda c/v \tag{21}$$

from the particle in transverse directions and has a size equal to the particle's de Broglie wavelength λ along the particle path; c is the velocity of inertons, which can be put here equal to the speed of light, and v is the velocity of the particle.

The transition to the orthodox abstract formalism of quantum mechanics is simple. And, in particular, submicroscopic mechanics accounts for the physical sense of the wave ψ-function attributing to it the inert properties of the particle. Therefore inertons are a substructure of the matter waves and the wave ψ-function represents a region of the space filled by the particle's inertons and thus the ψ-function formalism is correct only in the region covered by the particle's cloud of inertons.

In condensed matter the amplitude of the inerton cloud Λ (21) of a vibrating atom/molecule achieves a value of around 1 μm. The motion of any entity in the space obeys the laws of elasticity and, because of that, entities in matter are also exposed to the elastic interaction through their inerton clouds (see Ref. [22] for details). Hence the interaction

caused by the overlapping of inerton clouds of entities in matter must undoubtedly be taken into account and this kind of interaction is available even in gases. In fact, entities can contact each other until their concentration n satisfies the inequality

$$n^{-1/3} < \Lambda, \quad \text{or} \quad n < \left(hc/mv^2\right)^3 \tag{22}$$

where m and v are the mass and the velocity of a molecule in the gas studied. Depending on typical values of m and v the concentration of gas whose molecules suffer from the elastic inerton interaction can crudely be estimated as $n \approx 10^{12}$ to 10^{21} m^{-3}.

Coming back to the problem of clusters in water, we have right now to introduce a typical elastic interaction between water molecules, $\frac{1}{2} m\,\omega^2\,\delta g^2$, which has be added to the potential (20); here $\omega = 2\pi\nu$ and δg is the difference between displacements of two interacting molecules from their equilibrium positions, as is the case with particles in a crystal lattice. Such a kind of interaction is very important in solids, because it directly results in phonon spectra in the crystal lattice. (Recall once again that the inner reason of this interaction is stipulated from the elastic inerton interreaction of vibrating entities of the lattice and the value of displacement δg of an entity from its equilibrium position, which is about 0.01 nm, must be identified with its de Broglie wavelength λ in the crystal lattice.)

Since the water network is a kind of a lattice as well, the aforementioned vibratory spectra, i.e. elastic vibrations caused by the overlapping of inerton clouds of water molecules, should also be taken into account. Thus the respective attractive (U) and repulsive (V) paired potentials needed for the problem of clustering of water molecules become

$$U\left(xg,\ x\delta g\right) = \alpha \frac{e^2}{4\pi\,\varepsilon_0 \varepsilon\, g x} \ - \ \frac{1}{2}\, m\,\omega^2\,\delta g^2\, x^2, \tag{23}$$

$$V\left(gx\right) = V_0 \exp\ (-x)\,, \tag{24}$$

here we have introduce the dimensionless variable x; m is the mass of a H_2O molecule; $\omega = 2\pi\nu$ where ν is the eigenfrequency of collective behavior of water molecules in the system in question.

4 Cluster formation

With the use of expressions (23) and (24), the functions $a\,(\aleph)$ and $b\,(\aleph)$ determined in expression (18) become

$$a = \frac{3\,V_0}{k_{\rm B}T} \int\limits_1^{\aleph^{1/3}} e^{-(x-1)}\, x^2 dx \simeq \frac{3\,V_0}{k_{\rm B}T} \left(-x^2 - 2x - 2\right)\, e^{-(x-1)}\Big|_1^{\aleph^{1/3}}$$

$$\simeq \frac{15\,V_0}{k_{\rm B}T}, \tag{25}$$

$$b = \frac{3\,\alpha\,e^2}{4\pi\varepsilon_0 \varepsilon\, g\, k_{\rm B} T} \int\limits_1^{\aleph^{1/3}} \frac{1}{x}\, x^2 dx \ + \frac{3}{2} m\,\omega^2\,\delta g^2 \int\limits_1^{\aleph^{1/3}} x^2 x^2\, dx$$

$$\simeq \frac{3\,\alpha\,e^2}{8\pi\,\varepsilon_0\,\varepsilon\,g\,k_{\rm B}T}\,\aleph^{2/3} \;-\; \frac{3\,m\,\omega^2\,\delta g^2\,\aleph^{5/3}}{10\,k_{\rm B}T}. \tag{26}$$

If we substitute functions (25) and (26) in expression (21) for the action S, we obtain

$$S = \frac{30\,V_0}{k_{\rm B}T}\,\aleph \;-\; \frac{3\,\alpha\,e^2}{4\pi\varepsilon_0\,\varepsilon g\,k_{\rm B}T}\,\aleph^{5/3} + \frac{3}{10}\,\frac{m\,\omega^2\delta g^2}{k_{\rm B}T}\,\aleph^{7/3} + \;\aleph\ln\xi. \tag{27}$$

The equation for the minimum of the S, i.e. $\delta S/\delta\aleph = 0$, is the following

$$\frac{30\,V_0}{k_{\rm B}T} \;-\; \frac{5\,\alpha\,e^2}{4\,\pi\varepsilon_0\,\varepsilon\,g\,k_{\rm B}T}\,\aleph^{2/3} + \frac{7\,m\,\omega^2\,\delta g^2}{5\,k_{\rm B}T}\,\aleph^{4/3} + \ln\xi = 0. \tag{28}$$

Retaining the two middle terms, which are major ones, the solution to equation (28) is reduced to

$$\aleph \simeq \left\{\frac{25\,\alpha}{14\varepsilon}\;\frac{\frac{e^2}{4\,\pi\,\varepsilon_0\,g}}{\frac{1}{2}m\,\omega^2\,\delta g^2}\right\}^{3/2}. \tag{29}$$

Note once again that this solution has emerged as a result of the competition of the two non-uniformly scaled interaction potentials of different natures: the first kind of interaction is the pure Coulomb one and the second kind of interaction is the purely elastic one caused by the reciprocal action of inerton clouds of oscillating molecules.

In line with the rules of calculations of the mean statistical value of the square of the displacement (see rules of calculations of the Bose (phonon) operators, e.g. in Refs. [23–25])

$$\delta g^2 = \delta g_0^2\coth\left(\frac{h\nu}{2k_{\rm B}T}\right) \tag{30}$$

(indeed, in the case of phonons described by the Bose operators of creation $\hat b^\dagger_{\mathbf q}$ and annihilation $\hat b_{\mathbf q}$, the mean value is found by the expression

$$\frac{\exp(-\sum_{\mathbf q}\hbar\omega_{\mathbf q}\hat b^\dagger_{\mathbf q}\hat b_{\mathbf q})}{{\rm Tr}\exp(-\sum_{\mathbf q}\hbar\omega_{\mathbf q}\hat b^\dagger_{\mathbf q}\hat b_{\mathbf q})}\left(\hat b^\dagger_{\mathbf k}+\hat b_{-\mathbf k}\right)^2 = \frac{\exp(-\hbar\omega_{\mathbf k}\hat b^\dagger_{\mathbf k}\hat b_{\mathbf k})}{{\rm Tr}\exp(-\hbar\omega_{\mathbf k}\hat b^\dagger_{\mathbf k}\hat b_{\mathbf k})}\left(2\hat b^\dagger_{\mathbf k}\hat b_{\mathbf k}+1\right) = (2n_{\mathbf k}+1)$$

where $n_{\mathbf k}$ is the Plank function).

Then taking into account that $\frac{1}{2}m\,\omega^2\,\delta g_0^2 = h\nu$, expression (29) can be rewritten as follows

$$\aleph \simeq \left\{\frac{25\,\alpha}{14\varepsilon}\;\frac{\frac{e^2}{4\,\pi\,\varepsilon_0\,g}}{h\nu\coth(\frac{h\nu}{2k_{\rm B}T})}\right\}^{3/2}. \tag{31}$$

By expression (31), the increase in temperature diminishes the number of molecules $\aleph$ in a cluster. Let us assign numerical values to the parameters: the smallest distance between nearest molecules $g = 0.281$ nm; Madelung's constant $\alpha = 1.7$; the permittivity $\varepsilon = 4$ (Stobbe and Peschel [26] pointed out that the dielectric constant ε changes from 5 to 10 when the thickness of water layer varies from 1 to 12 nm). At room temperature and molecular frequency $\nu = 400$ cm^{-1} $(= 400\ {\rm cm}^{-1}\times 3\times 10^{10}\ {\rm cm/s}) = 1.2\times 10^{13}$ Hz the cotangent in expression (30) reaches unity. Substituting the aforementioned parameters into expression (30), we obtain: $\aleph \approx 890$. At a frequency $\nu = 1$ cm^{-1} $(= 1\ {\rm cm}^{-1}\times 3\times 10^{10}$ cm/s$) = 3\times 10^{10}$ Hz, the value for molecules in a cluster, $\aleph \approx 850$. These values of $\aleph$ are close to the experimental ones fixed in Refs. [8, 9].

5 Concluding remarks

Using the methods of statistical mechanics we have shown that the homogeneous water network is unstable and spontaneously disintegrates to the nonhomogeneous state (i.e. peculiar clusters), which can be treated as an ordinary state of liquid water. However, the investigation of the dynamics of clusters is beyond the approach described. The number $\aleph$ of water molecules which comprise a cluster is a function of several parameters, the main one of them being the frequency ν. Moreover, the solution to the problem of clustering strongly depends on the kind of paired potentials.

External field sources (electromagnetic and inerton radiation, ultrasound, and temperature) can also strongly influence the water system by changing the conditions of the cluster formation treated above. Consequently, the number of molecules $\aleph$ that enters a cluster will also vary.

Chaplin [27] presented an icosahedral cluster model; he argues that the structure of the water molecule should be fundamental at the arrangement of different cluster species. Smirnov [28] discusses the effect of water clusters in water activated by a molecular resonance technology; he believes that water clusters represent rather formations of long-range water molecular domains and this conclusion is in line with our previous results [3].

The shape of clusters, their proper frequencies, and how dispersed is the probability of finding a cluster of size $\aleph$ are very attractive problems. Their resolution will allow us to understand the organization of water in detail, because the formation of lager-scaled statistical (dynamical) domains seems to play a major role in the phenomenon of "memory" of water systems. These are indeed prima facie questions in molecular biology and alternative, or information medicine, which are topical problems of today.

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