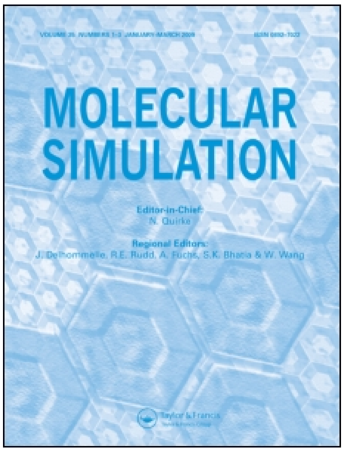

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Temperature dependence of the generalized frequency distribution of water molecules: comparison of experiments and molecular dynamics simulations

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Temperature dependence of the generalized frequency distribution of water molecules: comparison of experiments and molecular dynamics simulations

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The results of inelastic neutron scattering experiments on water in the temperature interval 300–623 K along the coexistence curve are compared with data obtained from molecular dynamics simulations. In general, a good agreement between experiments and calculations is observed and it serves as a satisfactory test of the potential models employed. The temperature dependence of the generalized frequency distribution of water molecules obtained by both experiment and computer simulation demonstrates the accordance with the temperature evolution of the water structure, extracted from neutron and X-ray diffraction measurements.

Keywords: Liquid water; Molecular dynamics; Inelastic neutron scattering; X-ray diffraction; Translational and librational motions

1. Introduction

Molecular motions in liquids may be experimentally studied by spectroscopic and scattering techniques (Raman, infrared, nuclear magnetic resonance, neutron scattering,...). However, the interpretation of the data from these experiments is a very difficult task, frequently based on approximate methods and acceptable but not absolutely rigorous hypotheses. In particular, the experimental mid-infrared frequency spectrum of liquid water (0–400 cm^{-1}) contains two marked broad bands around 60 and 170 cm^{-1}. These bands were observed from Raman spectroscopy measurements around 1930. More recently, Downing and Williams [1] detected the high frequency band in the experimental infrared spectrum of water and, a decade later, Hasted *et al.* [2] found the low frequency band, which shows a weaker signature in the infrared data. The interpretation of such features in terms of microscopic mechanical motions has been subject of discussion. From the observation of the weakening of both bands for temperature rise, the first interpretation of their meaning was due to the breakdown of the intermolecular structure. More specifically, Walrafen and co-workers [3,4] assigned the 60 cm^{-1} band to the bending of water molecules linked by hydrogen bonds and the 170 cm^{-1} band to the hydrogen bond stretching, although, they also indicated that both modes could be generally seen as restricted translations. Such interpretation in terms of frustrated translations is also supported by recent Raman spectroscopy measures [5].

From a theoretical point of view, analytical calculations and computer simulations have added relevant information helpful to enlighten the interpretation of both spectral modes [6–8]. However, in order to ensure the reliability of these theoretical results, the validity of the potential models employed has to be checked. This is usually a difficult task and, in general, it is only feasible when available experimental data can be compared with exactly the same physical quantities, obtained from analytical methods or from computer simulations.

The main objectives of the present paper are basically two: first, to perform a test of the validity of the flexible water model employed in a series of molecular dynamics (MD) simulations and second, to enlighten the

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interpretation of neutron and X-ray diffraction data in terms of microscopic motions with the help of the MD data. For such purposes, we have used fully corresponding results of frequency distributions in the range of translational and librational motions.

Several years ago, the group lead by Novikov carried out experiments on the study of the slow neutron interaction with water in a broad temperature range (from room till nearly critical, i.e. in the range 300–623 K along the coexistence curve) and performed the detailed analysis of those data. In particular, the temperature depending generalized frequency distribution (GFD) for water molecules (strictly speaking, for proton of water molecules) [9,10] was extracted. However, it was not possible to compare these experimental results with theory or MD simulations, because the absence of such data at that time. Some years later, this possibility arose: in the works [11,12] rose, MD simulation of the microscopic dynamics of liquid water was performed in broad range of the thermodynamic states, including those which were used in the experiments. Part of the computer simulation data, related with water spectroscopy at low frequencies, were not compared with experimental data and, consequently it was a lack of testing of the potential model employed.

The report given here is devoted to the comparison of the MD simulation data concerning the temperature dependence of GFD for water molecules with the experimental results. Such a comparison allows to estimate the validity of the MD calculation and to check the quality of the intermolecular potential used in it. In addition we will include, in the light of the comparison between both sets of data, some additional interpretation of the physical meaning of the results obtained.

2. The simulated GFD: computational details

The computational details of the MD simulations reported in this work are as follows: we considered 216 flexible simple point charge (SPC) water molecules at four states (298 K, 1 g cm^{-3}; 350 K, 0.97 g cm^{-3}; 403 K, 0.91 g cm^{-3} and 523 K, 0.75 g cm^{-3}) located along the liquid–vapor coexistence curve of the model, which does not coincide with that of real water [13]. The rigid SPC potential model of water consisted of three interaction sites located at the atomic positions [14]. We employed a flexible version which allows molecular bending and stretching vibrations, where such intramolecular motions were modeled according to the potential of Toukan and Rahman [15]. The suitability and efficiency of this flexible water model has been already shown [16]. However, we chose slightly different potential parameters in such a way that this reparameterized model reproduces satisfactorily the bands of the mid infrared spectrum of water [17]. Periodic boundary conditions and the Ewald sum rule to account for long-ranged Coulomb interactions were employed. A leap-frog Verlet integration algorithm with coupling to a thermal bath was used in all MD simulations [18]. In addition, molecular translational and internal degrees of freedom were separately equilibrated to reach the appropriate temperatures [19]. The integration time step was of 0.5 fs. The dynamical molecular properties were computed during the simulations in runs of more than 150 ps after equilibration runs of at least 20 ps for each thermodynamic state.

3. The experiment and data processing

The procedure of the inelastic neutron scattering experiment with the use of the double time-of-flight spectrometer DIN-2PI, as well as data processing and analysis applied were repeatedly described [20,21], and we shall not touch upon them here. It should be reminded only, that our approach to the analysis of inelastic neutron scattering results is based on the assumption that the inelastic neutron scattering spectra can be separated into two components: quasi-elastic and strictly inelastic. The first one contains the information, concerning with the diffusive motions of liquid particles, and we shall not discuss these questions here. The inelastic component carries the information about the vibration–rotation dynamics of a molecule in the force field of neighbors. If we deal with incoherent scattering, and it is the case for the neutrons of low energies scattered by water, the information about vibration–rotation MD is extracted in the form of GDF. In our experimental conditions it is the GFD for protons of a water molecule. From the physical point of view in the case of liquids, GFD can be understood as the frequency spectrum of the velocity autocorrelation function of atom given [22]. The procedure of GFD extraction from the results of inelastic incoherent neutron scattering experiment, elaborated and applied by our experimental group is fully described in Refs. [21,23].

4. The experimental results: comparison with MD simulation

The examples of GDF that we obtained are shown for two temperatures on figure 1. To provide the possibility for the quantitative estimation of GFD temperature deformation, we described the experimental curves by the simplified phenomenological model, which represents the experimental GFD at each temperature as the superposition of five Gaussian curves (the model of five non-interacting modes), each corresponding to the certain kind of molecular motion in the neighbors force field:

$$g(\varepsilon) = \sum_{i=1}^{5} C_i \left(\sqrt{2}\pi\sigma_i\right)^{-1} \exp\left[-\frac{(\varepsilon-\varepsilon_i)^2}{2\sigma_i^2}\right] \qquad (1)$$

where, C_i-weight coefficients (squares), ε_i-positions of maxima, σ_i-dispersions of the partial curves.

Two first low frequency modes in equation (1) correspond to the vibration (hindered translations) of water molecule, as the united dynamic unit with the mass of 18 a.u., and three high frequency modes relate to the hindered rotations (librations) of the water molecule around three main axis of inertia. The physical motivation for using such a model can be found in the results of MD simulation (figures 22 and 28 [24,25] and figure 1 [25]). Having obtained optimal description of the experimental curves by expression (1) on each temperature, we get the parameter temperature dependence for all five partial modes and then we have a possibility to describe quantitatively the GFD temperature deformations. The decomposition of the experimental GFD on the partial components is demonstrated for two temperatures by figure 1.

Before beginning the comparison, we should make two preliminary remarks. First, the calculations of the translational motion performed in reference [12] are presented in detail only for oxygen. But, since the mass of oxygen is close to the mass of the water molecule, and the molecular center of gravity for this molecule coincides approximately with the position of oxygen, we shall consider GFD of oxygen as the one corresponding to the translational motion of water molecule as a whole (as a common dynamic unit, including proton). Concerning the rotational part of GFD, the calculation for proton given in reference [12] just corresponds to that we get from the analysis of the neutron experiment. Second, as was mentioned above, our method of inelastic neutron spectra analysis includes the separation of these spectra into quasi-elastic and strictly inelastic parts. In GFD, we extract from experimental data, the diffusion modes are absent, whereas in the calculations they are included in the translational part of oxygen GFD.

For convenience of the comparison between experiment and simulation, we have described the simulated GFD by the same model (1), with the only difference that for accounting diffusion modes the translation part of GFD was separated into three components. In doing so, the diffusive mode has been approximated by an additional Lorentzian curve.

Experimental and calculated librational parts of GFD at two temperatures are shown in figure 2. In the case of MD

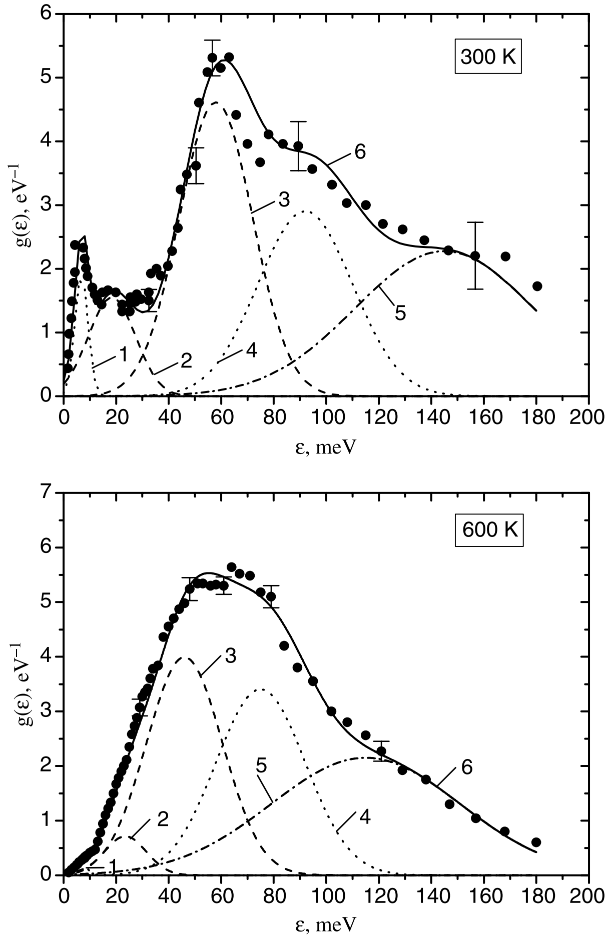


Figure 1. The decomposition of the experimental GFD on the partial components. ●, experimental results [10]; 1, first translational mode; 2, second translational mode; 3, first librational mode; 4, second librational mode; 5, third librational mode; 6, common approximating curve.

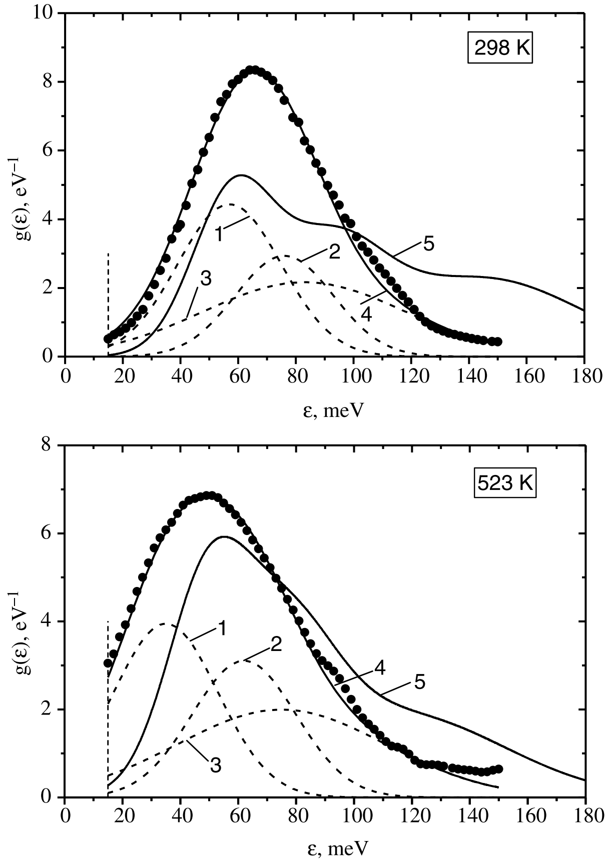


Figure 2. The librational part of the proton GFD in water molecule, obtained by the MD—simulation, decomposed on the partial components: ●, MD—simulation [12]; 1, first librational mode; 2, second librational mode; 3, third librational mode; 4, common approximating curve; 5, the librational component of the proton GFD, obtained experimentally [10].

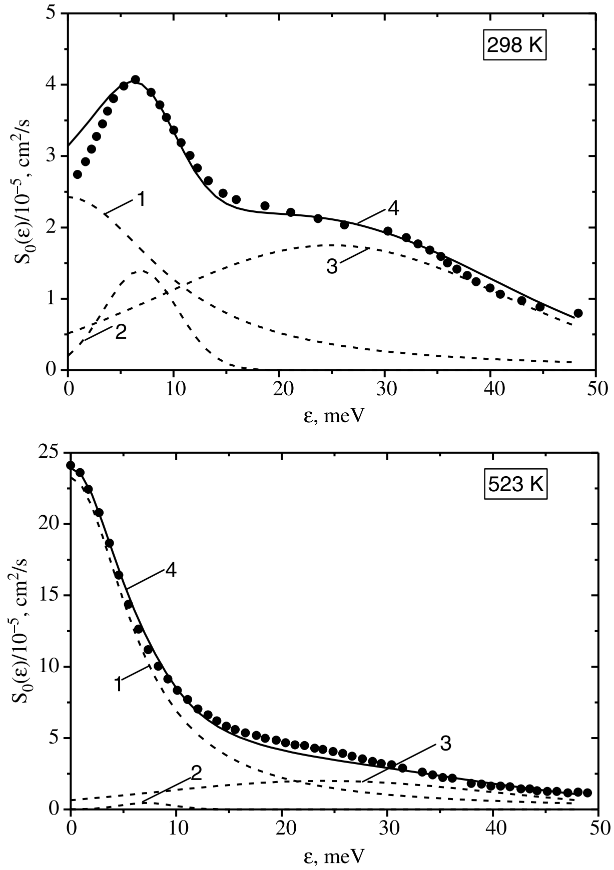


Figure 3. GFD of water molecule oxygen, obtained by MD—simulation, decomposed on the partial components: ●, MD—simulation [12]; 1, diffusive mode (lorentzian); 2, first translational mode; 3, second translational mode; 4, common approximating curve.

results the librational parts of GFD have been obtained by subtraction of the translation components from the corresponding common GFD curves.

The decomposition of the translational part of the calculated GFD on the partial components is shown in figure 3. The temperature dependence of the parameters for the discrete librational and translational modes obtained by both methods is shown in figures 4 and 5.

5. Discussion of the results

Inspection of figure 2 reveals that in the forms of the libration modes between the experimental and simulation results there exist observable differences. At the same time the average characteristics of these modes (positions, weights), with the exception of the third (high frequency) librational mode, manifest a remarkable agreement (figure 4). The same conclusion can be drawn from figure 5 relative to the translation modes, where the visible difference between the calculation and the experiment is observed only for the position of the second translational mode.

Concerning the GFD temperature dependence from the analysis of the calculated and experimental results it follows:

(1) The positions of librational modes (i.e. the force field in which the molecule librates) demonstrate some temperature decreasing only in the region of 300–400 K, remaining practically constant at higher temperatures; a similar temperature dependence displays the average force constant of the librational field (figure 6), calculated by the expression [10,26]:

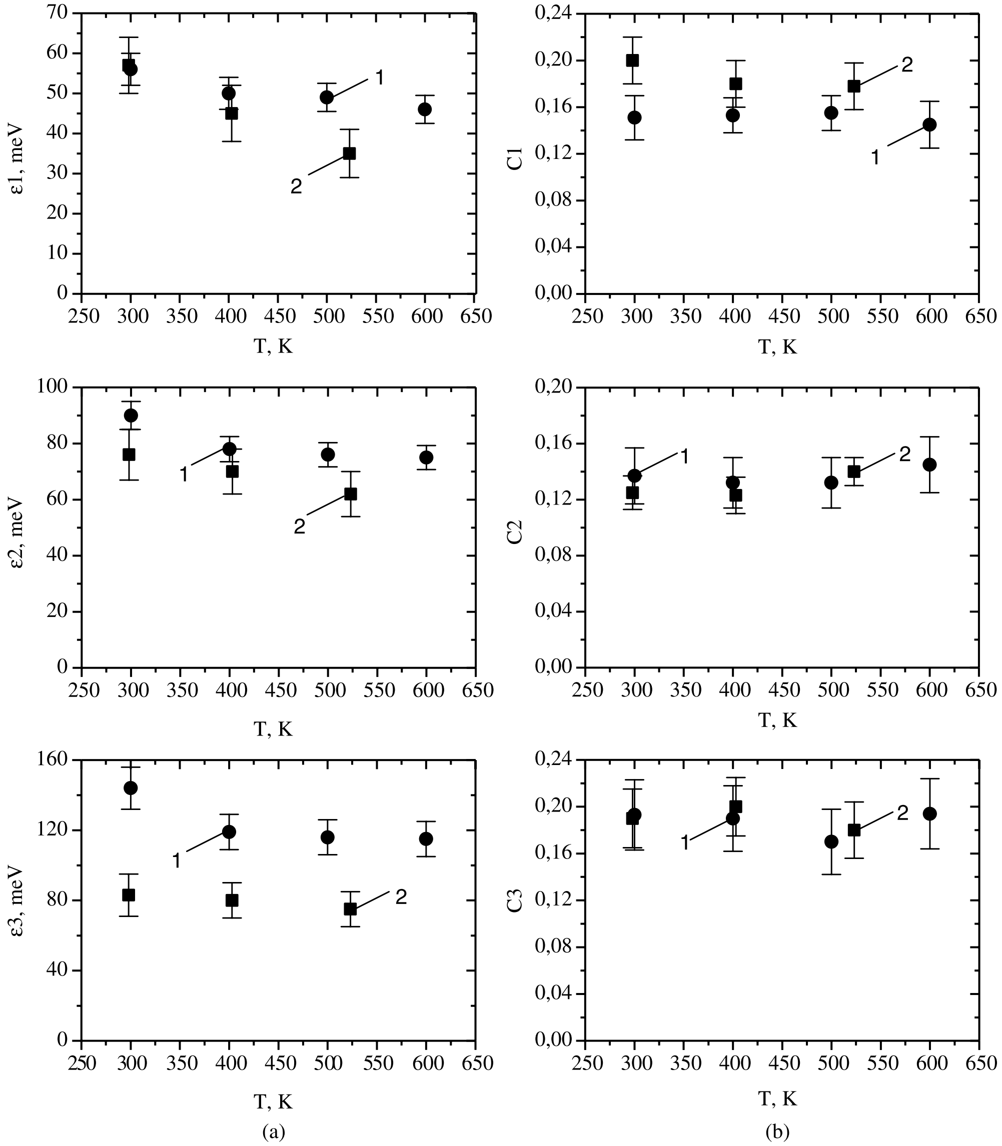
$$\langle \mathrm{K}_\theta \rangle(\mathrm{T}) = \frac{\bar{I}}{2\eta^2} \int_0^\infty \frac{\varepsilon^2 g_{\mathrm{lib}}(\varepsilon, T)\mathrm{d}\varepsilon}{(R-r)r} \qquad (2)$$

where, R is the intermolecular distance; r is the O–H distance in water molecule; g_{lib} is the librational component of the GFD and $\bar{I}$ is the mean moment of inertia of a molecule ($\bar{I} \sim 2 \times 10^{-40}\,\mathrm{g\,cm^2}$).

(2) The weights of the librational modes over the temperature region investigated remain constant (of course, in the limits of experimental errors). Since the librational modes have the nature of "local" excitations and are defined by nearest environment [27], the picture we get can be considered as an evidence of the temperature stability of the short range order in water.

(3) The most remarkable peculiarity in the temperature behavior of the translational modes is the quick decay of the weight of the first translational mode at elevated temperatures; this (lowest frequency) mode is understood usually to be connected with bending the hydrogen bond (HB) angle O···O–O [26], and its decay points to the disruption of the common space HB network in liquid water, existing in it under normal conditions; the force constant, responsible for this kind of molecular motion demonstrates the quick temperature decay, as well (see figure 7). It is interesting to remark, that the similar behavior of the first translation mode takes place in the hydration shell of ions in the aqueous solutions [28]. However, since this particular mode is also present in the spectra of non-associated liquids, we think that its microscopic interpretation could be assigned, from a more general point of view, to restricted translations occurring in all sorts of liquids [8].

It seems important to correlate the above data from microscopic dynamics with the information known currently about the structure of water and its temperature dependence. These questions have been actively studied for many years and continue to be studied today with use of X-ray diffraction [29,30], neutron diffraction [31–34] and MD simulation [35–37]. The temperature evolution of the water structure can be traced with figure 11 given in Ref. [34] (we have no room to show this picture), where the partial pair correlation functions $g_{\mathrm{OO}}(r)$, $g_{\mathrm{OH}}(r)$, and $g_{\mathrm{HH}}(r)$ obtained experimentally are presented. The results of this work reflect in general the modern point of view on the problem discussed. It follows from this drawing that spatial short-range order in



(a) (b)

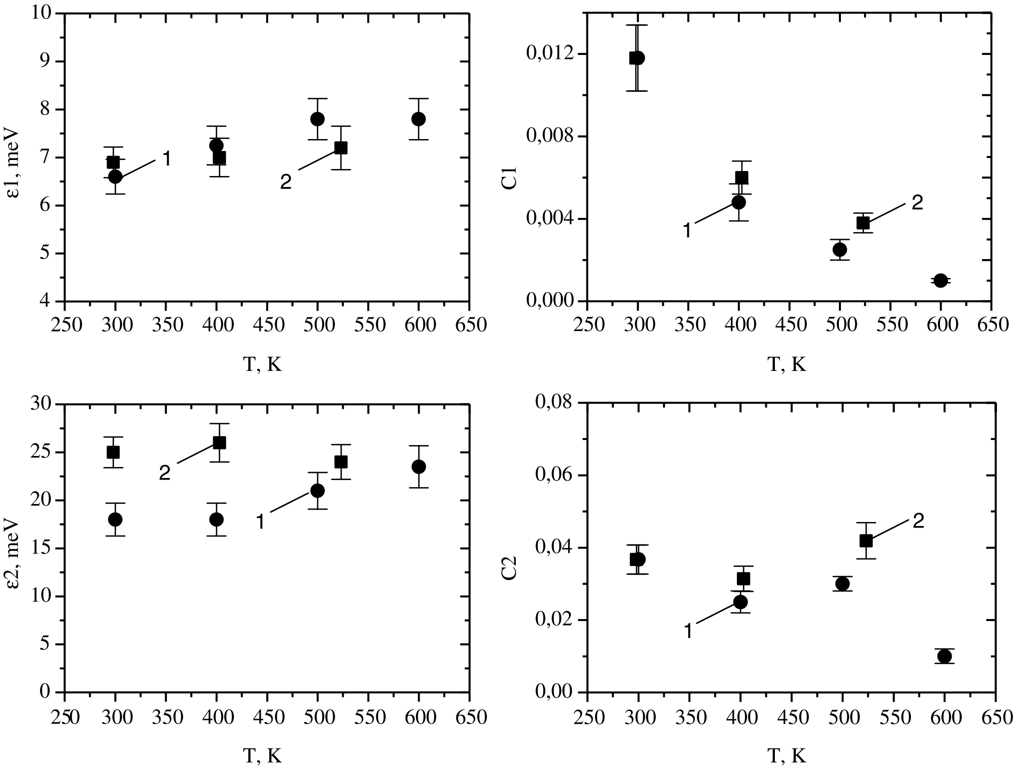
Figure 4. The temperature dependence of positions (a) and weights (b) of librational modes: 1, experimental results [10]; 2, MD—simulation [12].

liquid water demonstrates the significant temperature stability. This line of the water structure behavior is reflected in the temperature evolution of $g_{OO}(r)$ function and in particular by its first peak. At higher temperatures this peak decreases in amplitude, moves to the direction of bigger r but, however, remains clearly determined. It correlates with our conclusion about temperature stability of librational modes, which are conditioned by the local environment.

Orientational ordering in liquid water appears to be less stable with temperature. First, at the elevated temperatures the disappearance of the second peculiarity in $g_{OO}(r)$ function about $r \sim 4.5$ Å, corresponding to the second neighbors takes place. Second, the figure discussed shows the quick temperature deformation of the first peaks in $g_{OH}(r)$ and $g_{HH}(r)$ functions about (400–450 K). It is well known that these peaks reflect the existence of the space HB network in water and their temperature deformation indicates the decay of the common space HB network around these temperatures. In the terms of percolation model [38,39], in this temperature region water ceases to be the united percolation cluster, but is splitted into the number of smaller ones. Now the typical ordinary water tetrahedral coordination extends only over the close environment. This structure observation is in good agreement with our conclusion about the quick temperature decay of the first translational mode related to the presence in water of the extended space HB network.

6. Concluding remarks

As our final conclusions, we would like to underline two points. Firstly, from the common qualitative agreement of the experimental results and MD simulation, it follows



(a) (b)

Figure 5. The temperature dependence of positions (a) and weights (b) of translational modes: 1, experimental results, [10]; 2, MD—simulation [12].

that the effective pair intermolecular potential employed in Ref. [12] allows to reproduce the main features of the rotation-translation dynamics of water molecules in the intermolecular force field and the temperature evolution of this dynamics. In the light of above analysis, it would be interesting to try some additional reparameterization of this potential which could help to improve the agreement with the experiment. Secondly, from our analysis it follows that there exist evident correlations between the temperature dependence of the microscopic dynamics of liquid water, obtained experimentally and by MD simulation, and the contemporary understanding of the temperature evolution in the water structure. Generally speaking it is not a surprising fact, because the MD as well as the molecular structure of a liquid are determined by a principal common factor, i.e. the potential of intermolecular interaction.

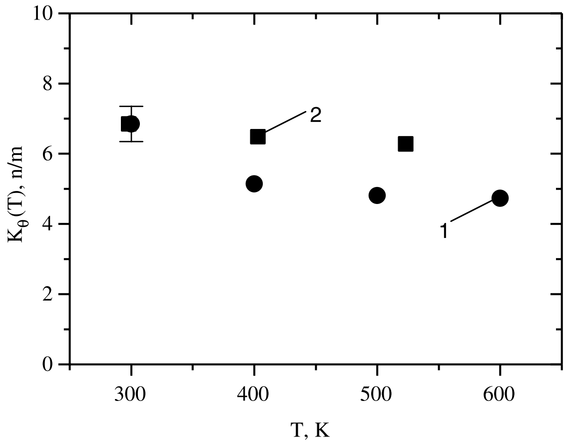


Figure 6. Force constants for rotational motions: 1, experimental results [10]; 2, MD—simulation [12].

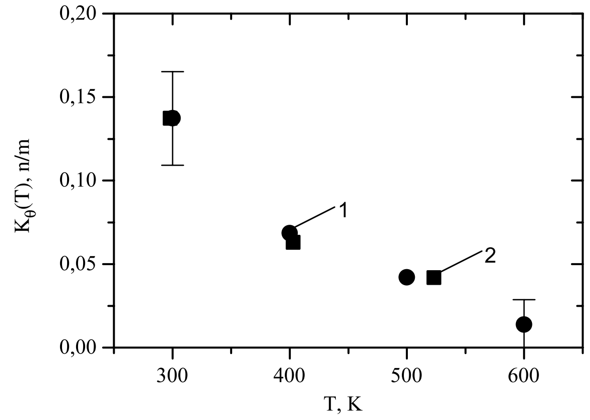


Figure 7. Force constants for translational motion: 1, experimental results (first translational mode) [10]; 2, MD—simulation (first translational mode) [12].

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