ISSN 0006-3509, Biophysics, 2014, Vol. 59, No. 6, pp. 973–985. © Pleiades Publishing, Inc., 2014.
Original Russian Text © A.V. Drozdov, T.P. Nagorskaya, 2014, published in Biofizika, 2014, Vol. 59, No. 6, pp. 1195–1208.

DISCUSSIONS

The Quasi-Periodic Character of Intermolecular Interactions in Water

A. V. Drozdov and T. P. Nagorskaya
Institute of Analytical Instrument Making, Russian Academy of Sciences, St. Petersburg, 190103 Russia
e-mail: av@biophys.ru

Received February 10, 2014; in final form, June 15, 2014

Abstract—The dynamics of physical characteristics of water are studied using various methods of molecular structural analysis: IR spectroscopy, Raman spectroscopy, microwave radiometry, and nuclear magnetic resonance in the magnetic field of the Earth. The changes in the physical characteristics of water obey certain regularities. Similar and well-reproducible oscillation periods of the measured values are observed in all experiments, regardless of the method of analysis used. These periods are 1–3, 5–9, 10–13, 14–18, 21–29, 30–39, 41–55, and ~60 min. The oscillation amplitudes vary up to 10%. Based on the two-structure model of water, the revealed quasi-periodic character of the intermolecular interactions may be related to the dynamics of mutual transitions between local structural inhomogeneities of water. It is proposed that the observed quasi-periodic character of the intermolecular interactions is due to spin isomerism of water molecules. The problems of the relationship between the oscillations of the physical properties of water and biorhythms are discussed.

Keywords: physical properties of water, structural peculiarities of water, dynamics of physicochemical properties of water, spin isomerism of water, ortho and para water molecules, intermolecular interactions in water, rhythmic processes, biorhythms

DOI: 10.1134/S0006350914060025

INTRODUCTION

Physicists, chemists, and biologists have been thinking for many decades about the possible role of water in biological processes. The unique role of water is not only that water serves as a specific medium for biochemical reactions, but may possibly also participate directly in the regulation of biological processes. Several researchers believe that it is water that creates prerequisites for controlling bioprocesses at the level of macromolecules of biopolymers, as well as at the membrane and cell levels [1–3]. Scientists all over the world are devoting much attention to the problem of the local "structure" of water [4–8]. (We put "structure" in quotation marks to separate this idea from the purely crystallographic interpretation of the exclusively static character. The idea of the "structure" of water implies the dynamic ordering as well.) It is assumed that the "structural" rearrangements of water under the action of external factors, including low-intensity factors, are responsible for many biological phenomena [2, 9].

However, in spite of the numerous works studying the "structure" of water [10, 11], the majority of the scientific community is still doubtful that there is a fundamental possibility of a substantial influence of the "structural" rearrangements on processes in living systems. This skepticism is based on the thermodynamic approach used by modern science in the study of the role of water in biological processes. This approach postulates that the state of the system cannot be changed by an external effect the energy of which is much lower than the thermal energy of the system (the value of which is about kT).

Nevertheless, a critical volume of experimental results indicating the influence of physical low-intensity factors on both biological systems and the physicochemical properties of water (see references cited in [12, 13]) has been accumulated to date in the scientific literature. However, many of these results are poorly reproducible. A paradoxical situation is observed: on the one hand, the accumulated facts indicate that the external physical fields affect the "structure" of water and, on the other hand, a poor reproducibility does not favor the idea that there is a fundamental possibility of a similar effect.

The attempt to solve this paradox assumes that the main reason for the divergence of the obtained results is associated with the poor reproducibility of the experimental conditions. It is assumed that these or other phenomena can be observed only under rigidly certain conditions. If all these conditions are not fulfilled, the results will not be reproducible completely or partially. Since the mechanisms of "structural" rearrangements in water remain almost unstudied to date, it is impossible to reveal what experimental conditions are not fulfilled.

We found a phenomenon that may also be a reason for the "poor" reproducibility of experimental results of the molecular structural analysis of water. The essence of the phenomenon is a quasi-harmonic change in the characteristics of a water system. The obtained results are not related to measurement errors and are caused, in our opinion, only by the dynamics of intermolecular interactions in water under the action of the external electromagnetic fields.

EXPERIMENTAL

Methods

Bidistilled water obtained in a glass distillator was used as samples in all studies. The electroconductivity of water was ~2–4 μS/cm, pH ~ 6.0. The temperature of the samples was maintained constant using a DC 30-K 15 programmed liquid circulation thermostat (Thermo Scientific Inc., United States). The accuracy of maintenance of a specified temperature was $\Delta t = \pm 0.1$°C. For decoupling of the analytical equipment from oscillations of the circuit voltage, all instruments were connected to a MegaLine 5000 continuous source with double conversion ($W_{\max}$ = 3500 W) and a nonlinear distortion coefficient of <1%.

The following methods were used: Raman spectroscopy, IR spectroscopy, microwave radiometry, and nuclear magnetic resonance (NMR) in the magnetic field of the Earth. These methods are commonly used for analysis of the molecular structure of matter and are widely employed in physicochemical studies of water and aqueous solutions.

IR spectroscopy. IR spectroscopy is a universal physical method applied to the study of structural peculiarities of various organic and inorganic compounds. The method provides information on stretching, bending, and libration vibrations of water molecules, thus characterizing intermolecular interactions as a whole.

Absorption spectra of water were obtained on an IRPrestige-21 FT-IR spectrometer (Shimadzu, Japan). The full frequency range of the spectrometer was 350–7800 cm^{-1}. The spectra were recorded with a resolution of 1 cm^{-1}. Both the accumulation mode and single scanning of the spectrum were used. In the accumulation mode, each experimental point was averaged over 20 measurements. In this case, the time of recording of the full spectrum did not exceed 30 s.

The following "water regions" from the total spectrum were analyzed: the component vibrations $\nu_2 + \nu_L$ at 2290–1950 cm^{-1} ($\nu_{\max}$ ~ 2130 cm^{-1}), the stretching vibrations ν_{OH} at 3720–2820 cm^{-1} ($\nu_{\max}$ ~ 3400 cm^{-1}), the stretching–bending vibrations $\nu_2 + \nu_{OH}$ at 5600–4540 cm^{-1} ($\bar{\nu}_{\max}$ ~ 5180 cm^{-1}), and the overtone bands of stretching vibrations $2\nu_{OH}$ at 7300–6000 cm^{-1} ($\bar{\nu}_{\max}$ ~ 6880 cm^{-1}).

The results obtained for each "water" band were similar.

Dismountable Si tubes were used for the work in the near-IR range, and the thickness of the sample layer was varied from 50 to 100 μm for the bands $\nu_2 + \nu_L + \nu_{OH}$. The attenuate total internal reflectance (ATIR) method was used to detect the spectrum of the $2\nu_{OH} + \nu_2 + \nu_{OH}$ band. The ATIR method is based on the absorption by the surface layer of the studied substance of the electromagnetic radiation emitted from a total internal reflection prism that is in optical contact with the studied surface. The tubes for studies by this method were made of Si or Se, and the thickness of the water layer was ~5 mm.

Raman spectroscopy. Raman spectra were recorded on a RamanFlex 400 spectrometer (PerkinElmer, United States). The power of the laser radiation source was 300 mW, the power transmitted to the sample was ≤100 mW, and $\lambda = 785$ nm. The spectral resolution was 4 cm^{-1}. A water sample was placed in a light-protected body. The duration of laser irradiation was 4 s, and the time of signal recording was 4 s. The sample volume was ~250 mL. The region from 3000 to 3500 cm^{-1} of the stretching band of water molecules ($\bar{\nu}_{\max}$ ~ 3400 cm^{-1}) was used in the analysis.

Microwave radiometry. Among the numerous methods for studying the structure of matter, methods that provide information about the properties of the studied medium without a physical action on the studied object are especially valuable. One of these methods is near-field microwave probing in the passive mode. Analysis of intrinsic radio radiations of the studied sample makes it possible to obtain information about processes that occur at the molecular level.

Our experiments were carried out on a Radiometr supersensitive microwave radiometer (Telemak, Saratov, Russia) [14, 15]. The microwave radiometer had the following technical characteristics: sample volume ~75 mL, central receiving frequency 1 GHz, the receiving band at a level of 0.5–25 MHz, and sensitivity ~0.3 K (<10^{-17} W). The receiving microwave stripline antenna was matched with water at a frequency of 1 GHz, and the receiving antenna was excited by the magnetic component of the radio wave, which decreased the influence of background electromagnetic radiations and increased the radio probing depth.

The studies were carried out in an isolated anti-interference box with electromagnetic background suppressing not lower than ~30 dV (at the receiving frequency).

The scheme of the experimental setup is shown in Fig. 1.

Nuclear magnetic resonance in the magnetic field of the Earth. There are numerous methods for studying molecular structures, but only the resolution of X-ray

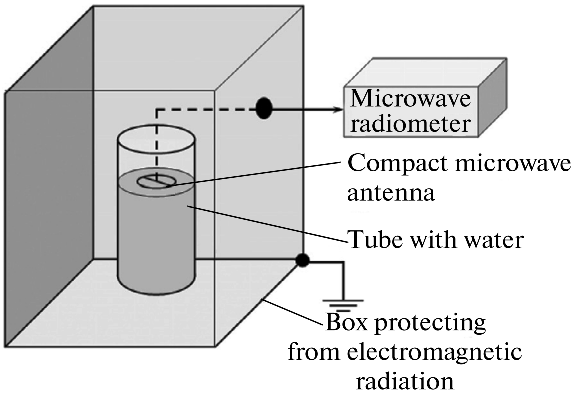


Fig. 1. Scheme of an experiment on microwave radiometry of water.

crystallography and NMR makes it possible to exactly determine the arrangement of atoms in a molecule, i.e., the molecular structure of compounds, as well as to recognize both static and mobile structures, providing information on the intra- and intermolecular dynamics.

Experiments on the observation of NMR in the magnetic field of the Earth were carried out at the Starorusskoe educational research station of the Department of Physics of the St. Petersburg State University (Russia). The operation of the experimental setup [16] is based on the principle of measuring the free nuclear induction [17].

Procedures

The time dependence of the intensity of the absorption or emission of the probing radiation by the water samples obtained using one of the methods described above was analyzed. This approach was chosen, because an increase or a decrease in the intensity level of the probing radiation is related to a change in the number of local structural inhomogeneities that absorbed or emitted the probing electromagnetic radiation (under the conditions of the constant volume of the sample and constant temperature, which was fulfilled in all experiments). In turn, the dynamics of local structural inhomogeneities is directly related to the dynamics of intermolecular interactions in the studied system.

The essence of all experiments was that the "water signal" was detected at equal time intervals (10 s, 30 s, 1 min, or 3 min) using one of the methods listed above. The duration of each experiment ranged from 60 to 600 min.

Procedures of IR spectroscopy and Raman spectroscopy. When IR spectroscopy and Raman spectroscopy were used for study, a spectral region containing the "water band" we are interested in was detected (Fig. 2a is the IR spectrum, and Fig. 3a is the Raman spectrum). The surface area under the curve in the earlier-specified frequency range of the "water band" (crosshatched regions) was calculated after recording of the spectrum to improve the signal to noise ratio. The integral intensity of the chosen spectral region was used in the further analysis. The chosen frequency ranges remained constant in all experiments.

Procedure of microwave radiometry. In the case of microwave radiometry, intrinsic radiations from water at a frequency of 1 GHz were measured continuously with a frequency of signal digitization of 10 Hz. To analyze the dynamics of the microwave radiation intensity, the whole signal was divided into 30-s intervals in the time scale and the power was calculated for these intervals. The power was the surface area under the curve, which was used in an analysis of the dynamics of the intensity of the microwave radiation from water (Fig. 4).

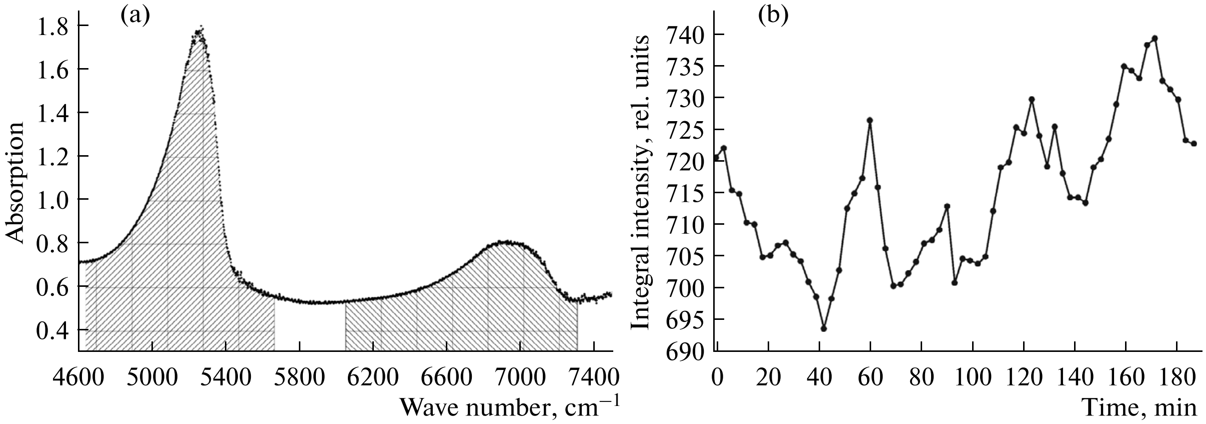


Fig. 2. (a) IR spectrum of the "water" bands ($\bar{\nu}_{max}$ ~ 5180 cm^{-1}, $\bar{\nu}_{max}$ ~ 6880 cm^{-1}) in the integration region (crosshatched) and (b) the dynamics of the integral intensity for the stretching–bending vibration band ($\bar{\nu}_{max}$ ~ 5180 cm^{-1}).

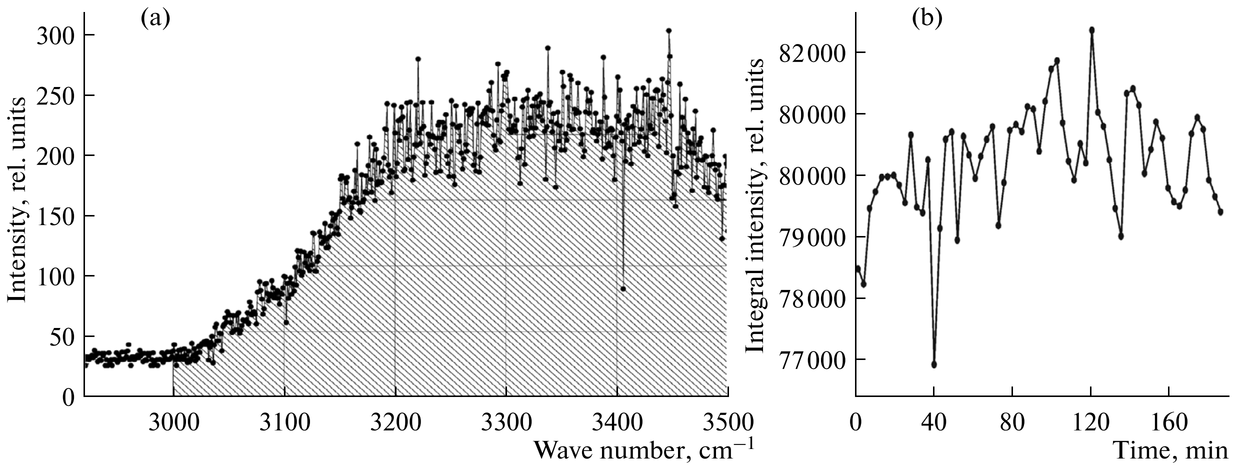


Fig. 3. (a) Raman spectrum and the integration region ($\bar{\nu}_{max}$ ~ 3400 cm^{-1}, crosshatched) and (b) the dynamics of the integral intensity for the stretching vibration band of water molecules ($\bar{\nu}_{max}$ ~ 3400 cm^{-1}).

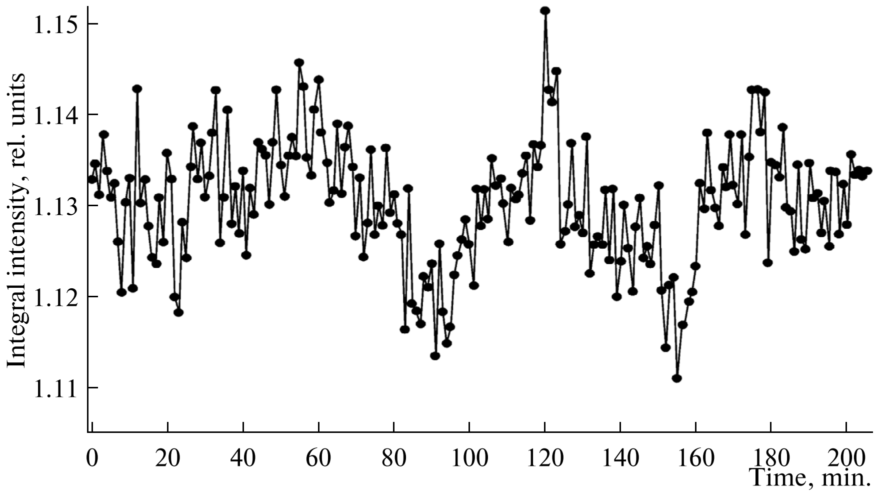


Fig. 4. Dynamics of the microwave radiation integral intensity at a frequency of 1 GHz.

Procedure of NMR measurements. The signal of a decrease in the free nuclear induction in the magnetic field of the Earth from protons [18] was analyzed in NMR experiments. The software of the NMR relaxometer used made it possible to obtain the spectrum of the signal (Fig. 5) and to "cut" the spectral region from the spectrum related to the response from protons only (crosshatched region in Fig. 5). The nuclear macroscopic magnetization was determined not by the amplitude of the central spectral line, but the integral intensity of the "proton" spectral region (crosshatched region in Fig. 5) was calculated. The dynamics of the integral intensity of the NMR signal from the water protons was used in the further analysis (Fig. 6).

The final stage of the study was frequency–time analysis of the integral intensity of the measured value (Figs. 2b, 3b, 4, and 6).

It should be mentioned that the results obtained in this work contained noises and were nonstationary in the time range. All this created difficulties for analysis of the obtained data by the accepted approaches (using the Fourier transform). Therefore, frequency–time analysis of the obtained experimental results was performed using the wavelet transform in the MATLAB 7.11 medium.

The wavelet transform was chosen because, unlike the Fourier transform, this transform makes it possible to analyze nonstationary processes and also gives a two-dimensional scan of the one-dimensional process at which the frequency and time are considered as independent variables. This makes it possible to analyze the properties of the studied process simultaneously in the time and frequency regions. The Morlet

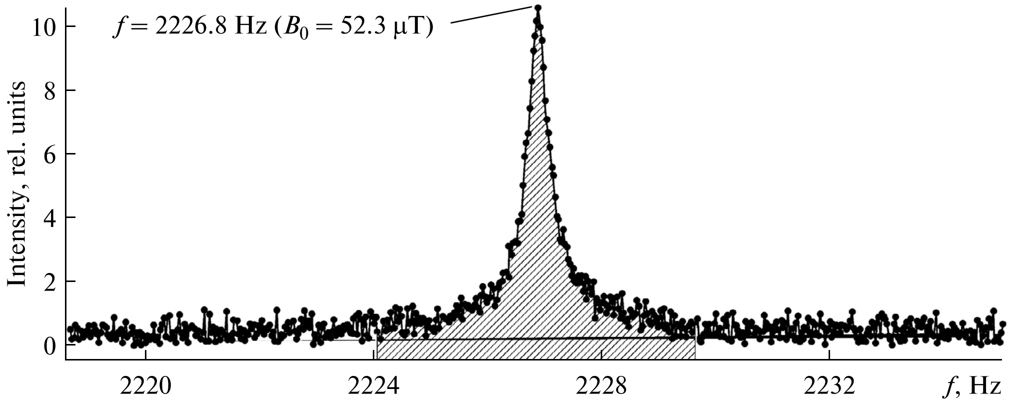


Fig. 5. Fourier spectrum of the decrease in the signal of the free nuclear induction in the magnetic field of the Earth (the spectral region of water protons is crosshatched).

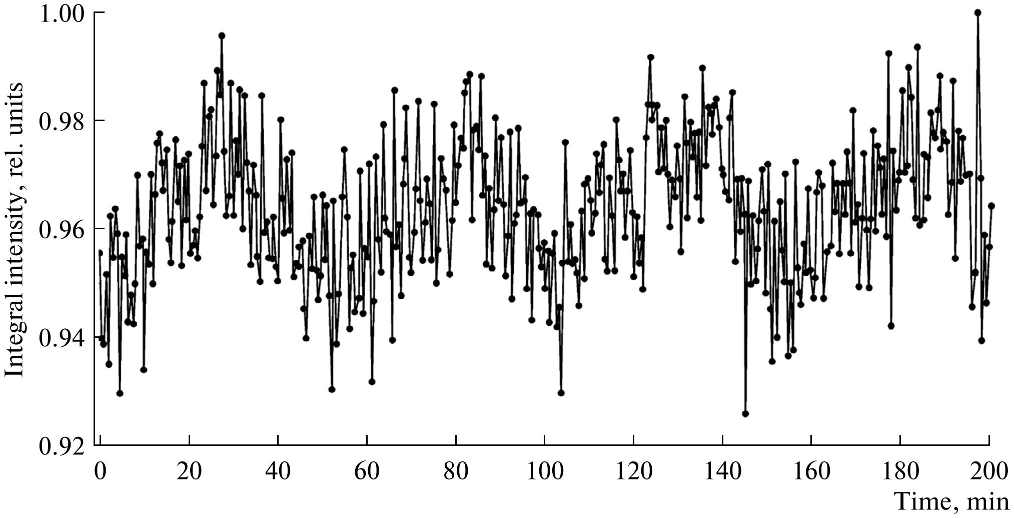


Fig. 6. Dynamics of the integral intensity ("proton density") of the NMR signal in the magnetic field of the Earth.

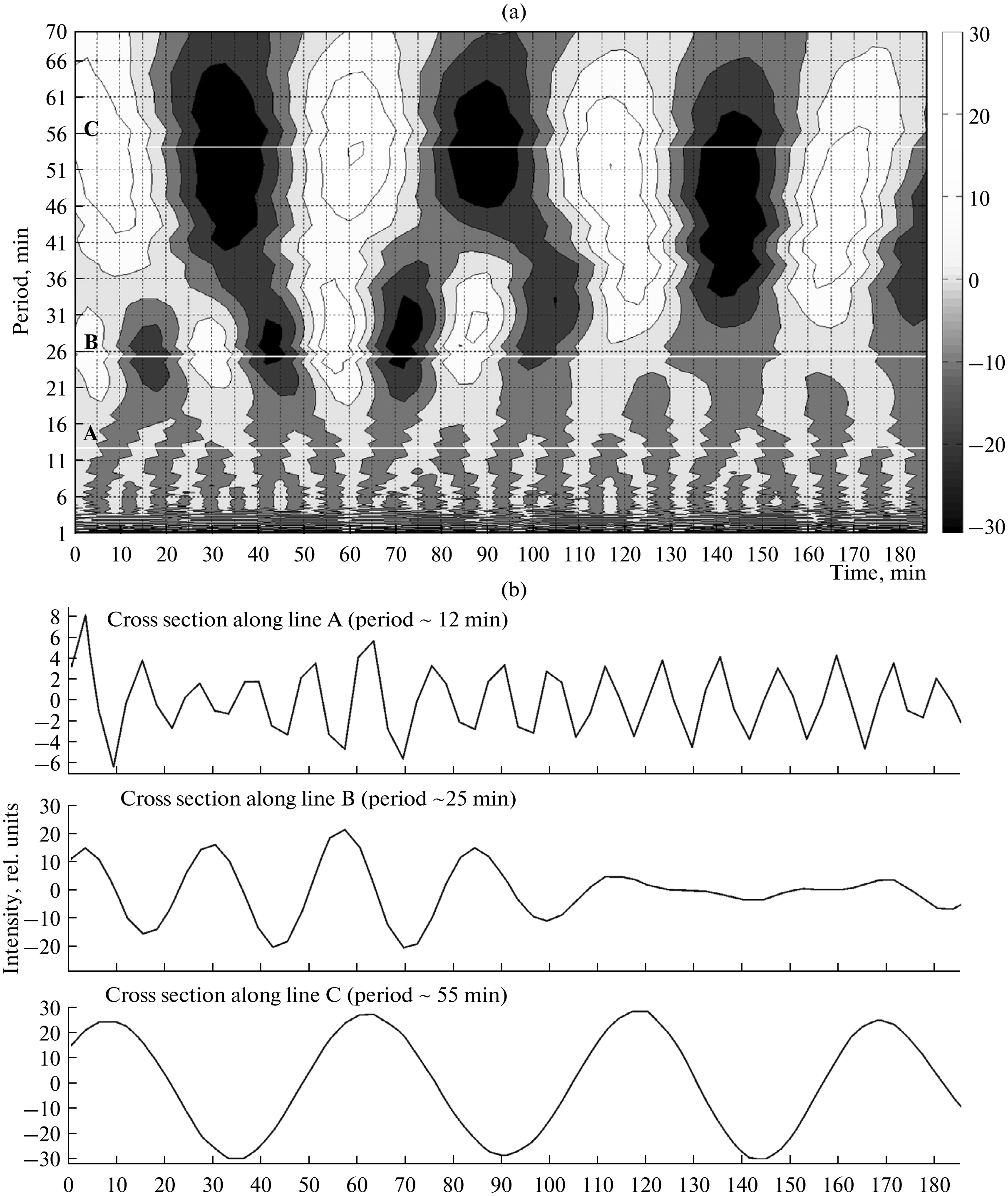
wavelet was used as a basis function of the wavelet transform.

Comparison of the results obtained by the wavelet transform with the results of the fast Fourier transform (for the most stationary regions of the experimental data) showed a good coincidence of the harmonic components obtained by both methods. The frequency of the sampling of experimental data detection did not affect the analysis results. All these facts indicate that the application of the wavelet transform is correct for analysis of the experimental results.

RESULTS AND DISCUSSION

Figures 7–10 show the results of the wavelet analysis of the dynamics of integral intensities of signals obtained by IR spectroscopy (Fig. 7), Raman spectroscopy (Fig. 8), microwave radiometry (Fig. 9), and NMR (Fig. 10). The presented results show that the dynamics of the absorption or emission intensities of the analytical signal contains similar harmonic components, regardless of the method used. An analysis of all experiments showed that the frequency spectra included the harmonics corresponding to periods of 1–3, 5–9, 10–13, 14–18, 21–29, 30–39, 41–55, and ~60 min. It is important that the obtained "water harmonics" are of high repetition, but it is not always the case that all harmonics are simultaneously present in the analyzed spectra.

It is well known that the results obtained by any spectroscopic method depend strongly on the smallest changes in the temperature of the sample because of the high sensitivity of these analytical methods. In our experiments, all samples were under temperature-controlled conditions. The amplitude of changes in the temperature of the sample did not exceed $\Delta t = \pm 0.1$°C in experiments on IR spectros-


Cross section along line A (period ~ 12 min)

Cross section along line B (period ~25 min)

Cross section along line C (period ~ 55 min)

Fig. 7. (a) Wavelet transform of the integral intensity dynamics for the stretching–bending vibration band in the IR spectrum of water $\bar{\nu}_{max}$ ~ 5180 cm^{-1} (see Fig. 2b) and (b) the harmonic components of the analyzed signal along the pitch periods.

copy, Raman spectroscopy, and microwave radiometry. In NMR experiments, $\Delta t = \pm 1°C$.

In spite of the good temperature maintenance, we carried out additional experiments, which made it possible to determine the degree of influence of the dynamics of the temperature of the sample on the dynamics of the measured physical values. In these experiments, the temperature of the samples was not maintained constant and the dynamics of their temperature was determined by the dynamics of the envi-

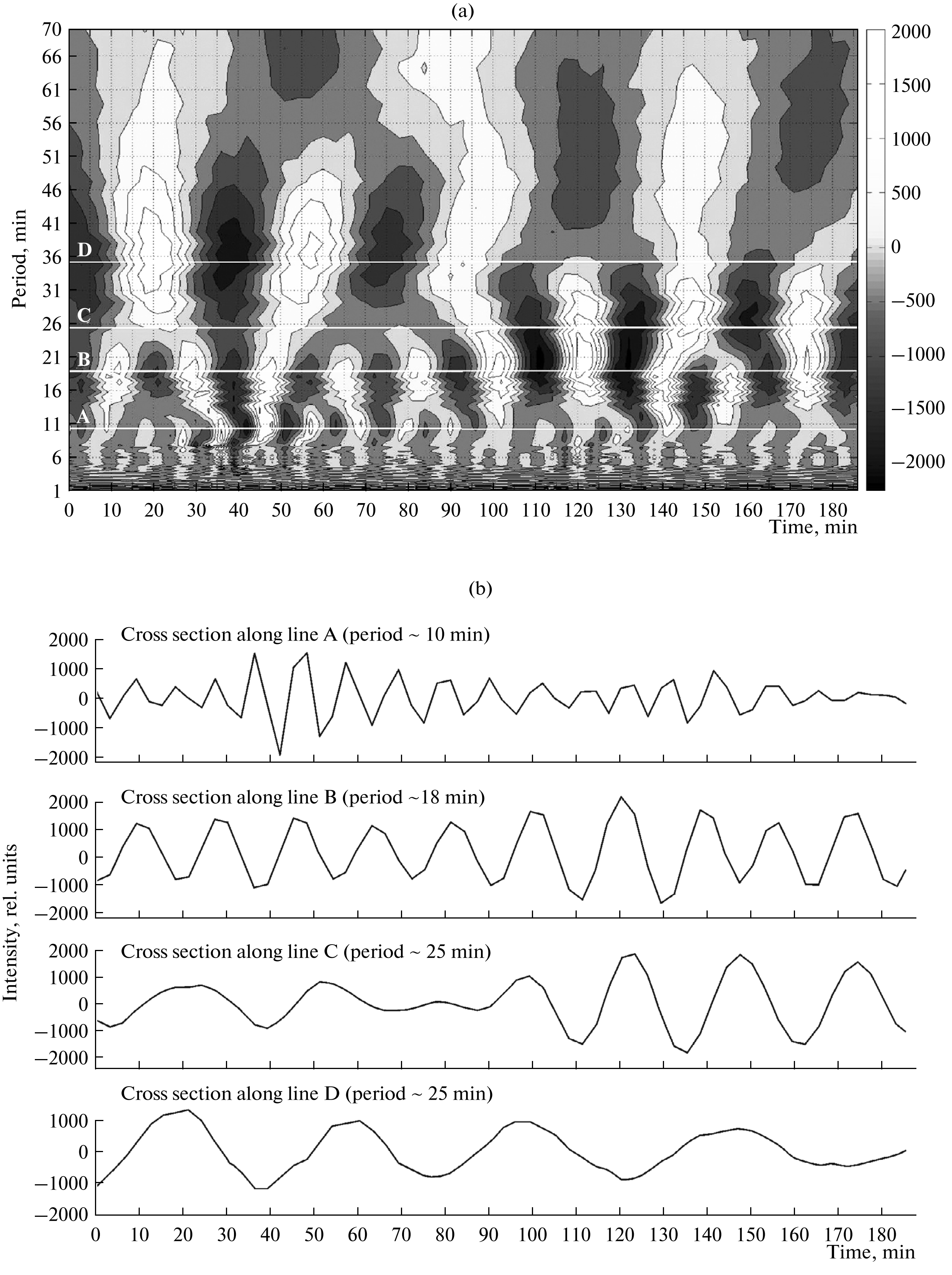


Fig. 8. (a) Wavelet transform of the integral intensity dynamics for the stretching band of water molecules $\bar{\nu}_{\max}$ ~ 3400 cm^{-1} in the Raman spectrum (Fig. 3b) and (b) the harmonic components of the analyzed signal along the pitch periods.

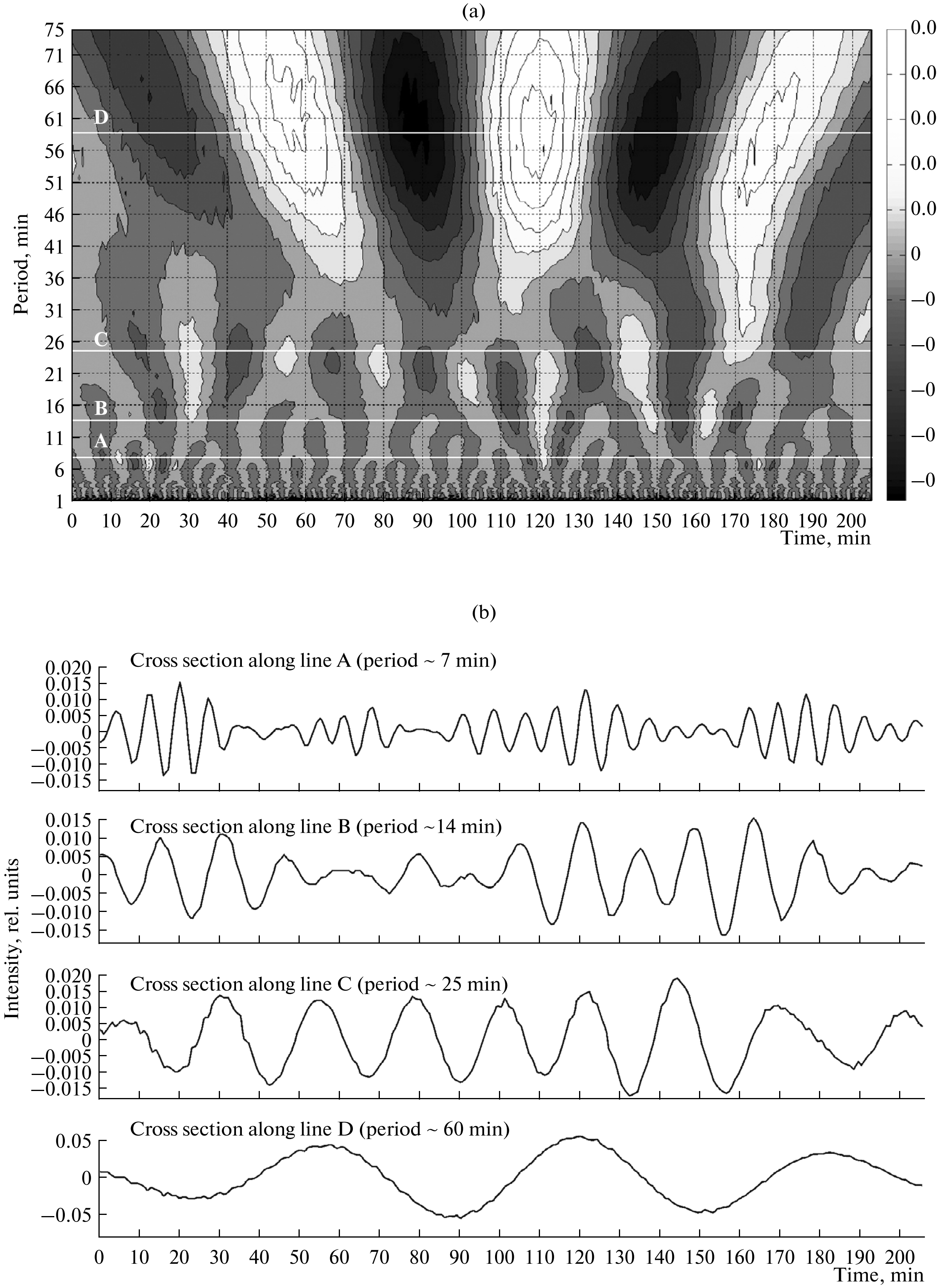


Fig. 9. (a) Wavelet transform of the dynamics of the microwave signal integral intensity from water at a frequency of 1 GHz (Fig. 4) and (b) the harmonic components of the analyzed signal along the pitch periods.

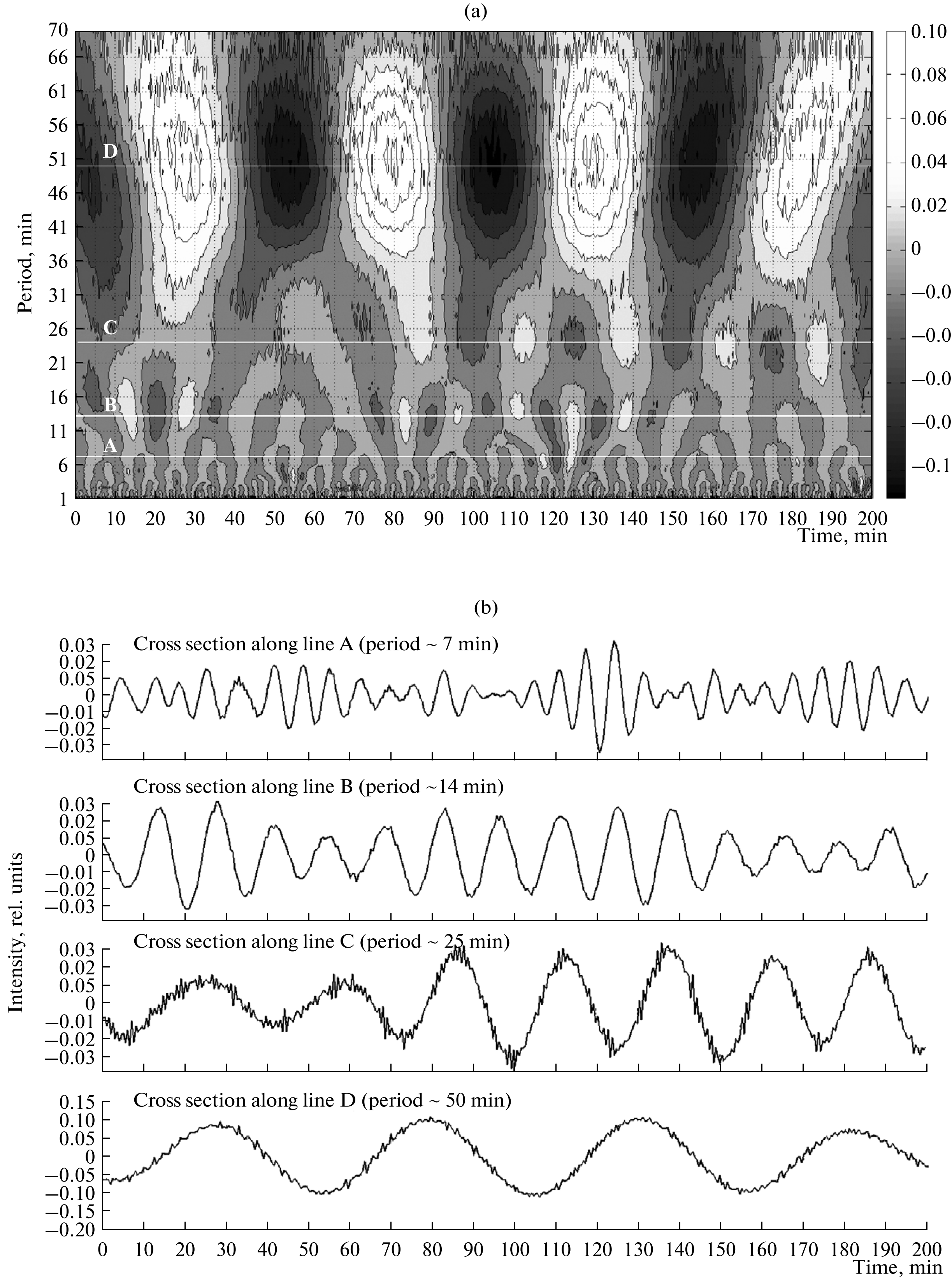


Fig. 10. (a) Wavelet transform of the "proton density" dynamics (the integral intensity of the NMR signal from water protons) in Fig. 7 and (b) the harmonic components of the analyzed signal along the pitch periods.

ronment temperature (the environment temperature was detected once a second). The obtained results indicate that the temperature of the sample does not determine the dynamics of the intensity of the absorption or emission of the probing radiation by water.

A good coincidence of the periods of the harmonic components in the dynamics of the measured values (obtained by different physical methods) can indicate that the observed quasi-harmonic character concerns the physical processes that occur in water at the molecular level. It is commonly accepted that water in the liquid state is a nonequilibrium system. Only the local equilibrium can exist in water, and its essence is the quasi-equality of continual transitions between the discrete states of water. Thus, the observed quasi-periodic processes reflect the dynamics of intermolecular interactions in water, i.e., the dynamics of mutual transitions between the local structural inhomogeneities of water. They are not related to experimental errors and peculiarities of the used methods of molecular structural analysis.

The results of Chernikov's works [19, 20] also indicate that the observed oscillations are not related to technical and methodological errors. Chernikov observed for the first time the oscillations of the light scattering intensity in water and in aqueous solutions using the dynamic light scattering method [19, 20]. Similar oscillations were observed [21] for the luminescence intensity in aqueous solutions. In all works, the oscillation periods well coincide with the harmonic components that we obtained.

To explain the obtained results, Chernikov suggested that *"according to the theory of molecular light scattering in liquid media, scattering occurs on optical inhomogeneities related to fluctuations of the temperature and density in the local regions of the medium. According to the two-structure models, the main concepts of which are retained in more adequate continual models, water has local differences in structure with a constant dynamics of mutual transitions (flickering clusters). Based on this, an increase and a decrease in the level of the scattered signal can be attributed to an increase and a decrease in the number of structural inhomogeneities, and the time ordering of similar structural transitions is shown in our work"* [19].

We believe that our results argue in favor of the hypothesis that local structural arrangements in water have a quasi-harmonic character. However, it remains quite unclear what reason or reasons can be responsible for the local "structural" rearrangements in water with times that are not characteristic of the relaxation processes in water.

Such methods of molecular structural analysis as IR spectroscopy, Raman spectroscopy, and microwave radiometry are highly sensitive, and the results obtained by these methods make it possible to *state* the quasi-harmonic character of the intermolecular processes in water. However, although these methods are precise and sensitive, they do not indicate any possible mechanism responsible for this behavior of the physical properties of water. In our opinion, the results of our NMR experiments can cast light on this problem.

The amplitude of the NMR signal from the sample is directly proportional to its nuclear macroscopic magnetization. According to the theory (see, e.g., [18]), the total nuclear magnetization in a constant magnetic field is the following:

$$M_0 = \frac{I+1}{I}\frac{N\mu^2}{3kT}B_0, \tag{1}$$

where I is the nuclear spin, N is the number of magnetic nuclei (in our experiment, the hydrogen nuclei of water molecules), μ is the magnetic moment of hydrogen nuclei, k is the Boltzmann constant, T is temperature (in absolute units), and B_0 is the external magnetic field induction.

It follows from an analysis of Eq. (1) that the integral intensity of the NMR signal (or, which is the same, "proton density") can change only upon a change in the number of magnetic nuclei or temperature of the sample. Let us consider the results of our NMR experiments from this point of view.

In all NMR experiments, the number of hydrogen nuclei remains unchanged, since the sample with water represented a hermetically closed vessel with a constant volume. The temperature oscillations in the room where experiments were carried out did not exceed $\Delta t = \pm 1°C$; i.e., the possible total amplitude of the temperature changes in the sample could not exceed 2°C. This value is less than 1% of the temperature changes in absolute units, as is required by Eq. (1).

The last factor that can affect the value of "proton density" is variations of the external magnetic field strength B_0.

The emission and absorption of the electromagnetic energy upon the change in the orientation of the magnetic dipole in the field B_0 occur at the frequency

$$\nu_0 = \frac{\Delta E}{\hbar} = \frac{\gamma}{2\pi}B_0, \tag{2}$$

where $\hbar$ is Planck's constant and γ is the gyromagnetic constant (in our case, for hydrogen nuclei).

From Eq. (2), we have

$$B_0 = \frac{2\pi\nu_0}{\gamma}. \tag{3}$$

Thus, the value of the external magnetic field was determined from the nuclear precession frequency using the same experimental results as those used for the determination of the integral intensity of the NMR signal (Fig. 6). It follows from an analysis of the NMR data that the variations of the external magnetic field strength (in our case, the magnetic field of the Earth) were lower than 0.1%.

In our experiments, the complete span of variability of the integral intensity of the NMR signal ("proton density") reached approximately 10% (Fig. 6). Thus, there is an explicit discrepancy of the observed change in the "proton density" and the explanations made in the framework of the existing theory [17, 18].

In fact, according to Eq. (1), the highest changeability in the NMR signal intensity may be due only to a change in the temperature of the sample, because the variability of the external magnetic field strength is more than an order of magnitude lower than the temperature variability. In turn, the value of the possible change in the temperature of the sample in NMR experiments is more than an order of magnitude lower than the value of the "proton density."

The discrepancies observed between the theory and practice can be explained if we assume that, in Eq. (1), the value of N (the number of magnetic nuclei in the sample) is not constant even for the sample with a constant volume. Let us refine this assumption.

When the number of magnetic nuclei involved in the NMR "response" is discussed, it is accepted that all nuclei present in the sample (in our case, protons) make a contribution to the NMR signal. However, this is not possibly so. For example, it is well known that "if hydrogen molecules are studied, an NMR signal is observed only from *ortho-hydrogen* (the total spin of which is $I = 1$)" [18].

Water molecules, as well as hydrogen molecules, can be either ortho (the spin of protons are oriented to the same direction, the total spin is $I = 1$), or para isomers (the spins of protons are oriented to different directions, and the total spin is $I = 0$). This fact allows us to assume that only protons with a certain spin orientation contribute to the NMR signal for both water and hydrogen molecules.

Thus, in terms of the assumption, the observed quasi-harmonic character of the integral intensity of the NMR signal from water may be due to the deviation from the equilibrium ratio between protons with different spin orientations in the whole volume of the sample. The total number of protons in the sample is undoubtedly unchanged. Equation (1) can be refined as follows:

$$M_0 = \frac{I+1}{I}\frac{N'\mu^2}{3kT}B_0, \tag{4}$$

where N' is the number of protons contributing to the NMR signal.

Consideration of the physical mechanisms responsible for the deviation of the ortho/para ratio of molecules from the equilibrium one is beyond the scope of this work and, hence, will be discussed elsewhere. Therefore, only some remarks on the topic will be made here.

Quantum transitions between different states of molecules should be synchronized in order to observe the overall oscillation effect of intra- and intermolecular transformations in the molecular system. First of all, one should attain intramolecular transformations to occur simultaneously (inphase transformations) in all molecules of the medium (or in the majority of them). On the one hand, the quantum transitions in molecules must be coherent to a sufficiently high extent. On the other hand, a resonance between the states corresponding to the basic energy levels is needed for manifestation of the periodicity effect, which is poorly possible under real conditions, because all levels of the systems are in the excited state. However, the periodicity effect can also be observed for the resonance of excited states but under a constant external effect. This may be indicated by our preliminary results that there exists a relationship between the oscillations of the physical properties of water and the variations of the external low-intensity electromagnetic fields. These results will be discussed elsewhere.

Our assumption about the relationship of the dynamics of intermolecular interactions in water and the dynamics of spin isomerism of water provides a new viewpoint concerning the "structure" of water.

Ortho and para water molecules differ by the rotational degrees of freedom (for the para molecules $J = 0, 2, 4, \ldots$, whereas for the ortho molecules $J = 1, 3, 5, \ldots$), resulting in the different characters of their interactions both with each other and in clusters. The possibility of incorporating water molecules into this or another structure depends on its rotational state [22]. Finally, the water "structure" is permanently rearranged in water. Therefore, it is not quite correct to speak of the "structure" of water in the crystallographic, static aspect. It is necessary to consider periodic changes in the "structure" itself. In turn, it can be assumed that the periodic external effects would affect the character of intermolecular interactions in water, thus determining the dynamic "structure" of water.

Our results are quite consistent with the idea [23] that water is a mixture of two liquids consisting of ortho and para water molecules.

The observed oscillation processes may cast light on our understanding of the mechanism of biorhythms, since it is water where biochemical reactions occur and form the basis for all living beings.

There is no commonly accepted theory of how the "biological clock" functions. Three major molecular biochemical hypotheses are mainly discussed in scientific schools: (1) auto-oscillations of biochemical processes, (2) genetic regulation, and (3) auto-oscillations of membrane permeability. Original work [24], in which the idea that spin conversion between ortho and para water molecules on the biological rhythms plays a determinative role was proposed for the first time as a mechanism of the "biological clock," should be emphasized.

In our opinion, the oscillation periods in water revealed in the present work, the rhythms found in living systems [25–29], and the good consistency of these values with the rhythms of space weather (periods of solar pulsations in the minute range are ~12, ~17, ~27, ~32, ~45, and ~57 min [30]) evidence the important role of water in the transmission of physical signals to biological objects. Recent works [31, 32] are especially worth mentioning in which the geomagnetic variations synchronous with the dynamics of the human physiological parameters were experimentally observed in the same frequency range as in our experiments with water. All these results indicate that the cosmophysical rhythm that may act in an evolutionary respect as a leading synchronization factor of biological processes.

To conclude, it can be thought that biorhythms may be based on the periodicity of physicochemical processes, which is tightly related to the quasi-harmonic character of intermolecular interactions in water. In our opinion, the character of interactions of water molecules (or its individual parts—for example, protons) with the external electric and magnetic fields is an important factor determining the formation of stable oscillation processes in aqueous solutions.

The effect of the magnetic field directly penetrating into the level of each cell can be directed to any of the magnetic moments in a biological system: the electron spin of the free radical, the nuclear magnetic moment, the orbital magnetic moment, the magnetic moment of the ortho water molecule, etc. These magnetic moments become unique “antennas” inside biological objects that can perceive the controlling effect from the magnetic field of the Earth and its variations [9] and can be a reason for the oscillation processes in water that we observed.

CONCLUSIONS

(1) The dynamics of changes in the physical properties of water is quasi-harmonic in character.

(2) The quasi-harmonic character of the physical properties of water may be a cause of partially or completely nonreproducible results in experiments on studying the influence of the external low-intensity effects on water.

(3) Analysis of the obtained results suggests that the observed dynamics of the physicochemical properties of H_2O is due to the dynamics of intermolecular interactions in water.

(4) It is not quite correct to say that water has a “structure” in the crystallographic meaning. The “structure” of H_2O permanently changes under the action of external factors. The *dynamic ordering of intermolecular interactions* can be observed at a certain harmonicity of the external effects in water.

(5) When discussing a possible mechanism responsible for the observed dynamics of intermolecular interactions, it is necessary to take into account spin isomerism of water molecules and the influence of the external electric and magnetic fields on the probability of spin conversion between H_2O isomers.

ACKNOWLEDGMENTS

We are grateful to E.V. Travkina, V.I. Pirushkin, and P.A. Kupriyanov for help in experiments and to S.M. Pershin, B.M. Vladimirskii, and P.M. Nagorskii for stimulating discussions.

This work was supported in part by the Presidium of the Russian Academy of Sciences, program no. 28 (subprogram “Physics, Chemistry, and Biology of Water”).

REFERENCES

1. A. Szent-Györgyi, *Bioenergetics* (Academic Press, New York, 1957; Fizmargiz, Moscow, 1960).
2. G. N. Ling, *Life at the Cell and Below-Cell Level: The Hidden History of a Fundamental Revolution in Biology* (Pacific Pres, New York, 2001; Nauka, St. Petersburg, 2008).
3. G. H. Pollack, *Cells, Gels and the Engines of Life: A New, Unifying Approach to Cell Function* (Ebner & Sons, Seattle, 2001).
4. W. C. Rontgen, Ann. Phys. Chem. N.F. **XLV**, 91 (1891).
5. O. Ya. Samoilov, *Structure of Aqueous Electrolyte Solutions and Hydration of Ions* (Akad. Nauk SSSR, Moscow, 1957) [in Russian].
6. L. Pauling, in *Hydrogen Bonding*, Ed. by D. Hadzi and H.W. Thompson (Pergamon Press, London, 1959), pp. 1–6.
7. G. N. Satsepina, *Physical Properties and Structure of Water* (Mosk. Gos. Univ., Moscow, 1998) [in Russian].
8. D. Eisenberg and W. Kautzmann, *The Structure and Properties of Water* (Oxford Univ. Press, New York, 1969; Gidrometeoizdat, Leningrad, 1975).
9. A. V. Drozdov, T. P. Nagorskaya, S. V. Masyukevich, and E. S. Gorshkov, *Biophysics* (Moscow) **55** (4), 652 (2010).
10. A. Nilsson and L. G. M. Pettersson, Chem. Phys. **389**, 1 (2011).
11. G. N. I. Clark, G. L. Hura, J. Teixeira, et al., Proc. Natl. Acad. Sci. U. S. A. **107** (32), 14003 (2010).
12. V. N. Binhi, *Principles of Electromagnetic Biophysics* (Fizmatlit, Moscow, 2011) [in Russian].
13. V. N. Binhi, *Magnetobiology: Underlying Physical Problems* (MILTA, Moscow, 2002; Academic Press, San Diego, CA, 2002).
14. V. I. Petrosyan, Yu. V. Gulyaev, E. A. Zhiteneva, et al., Radiotekhnika Elektronika **40** (1), 127 (1995).
15. V. I. Petrosyan, N. I. Sinitsyn, V. A. Elkin, et al., Elektron. Prom., No. 1, 99 (2000).
16. P. M. Borodin, A. V. Mel’nikov, A. A. Morozov, et al., *Earth’s Field Nuclear Magnetic Resonance* (Leningr. Gos. Univ., Leningrad, 1967) [in Russian].
17. M. E. Packard and R. H. Varian, Phys. Rev. **93**, 941 (1954).

18. *Quantum Radiophysics: Magnetic Resonance and Its Applications*, Ed. by V.I. Chizhik (St.-Peterb. Gos. Univ., St. Petersburg, 2009) [in Russian].
19. F. R. Chernikov, Biofizika **31** (4), 695 (1986). www.biophys.ru/archive/h2o-00005.pdf.
20. F. R. Chernikov, Biofizika **35** (5), 711 (1990). www.biophys.ru/archive/h2o-00006.pdf.
21. S. V. Gudkov, V. I. Bruskov, M. E. Astashev, et al., J. Phys. Chem. B **115** (23), 7693 (2011).
22. S. Ozeki and I. Otsuka, J. Phys. Chem. B **110** (41), 20067 (2006).
23. S. M. Pershin, Phys. Wave Phenom. **13** (4), 192 (2005). www.biophys.ru/archive/h2o-00002pdf.
24. D. I. Morré, P. J. Chueh, J. Pletcher, et al., Biochemistry **40**, 11941 (2002).
25. T. K. Breus and A. A. Konradov, *Effects of Solar Activity Rhythms: An Atlas of Temporal Variations in Natural, Anthropogenic, and Social Processes* (Yanus-K, Moscow, 2002), vol. 3 [in Russian].
26. V. S. Martynyuk, B. M. Vladimirskii, and N. A. Temur'yants, Geofiz. Protsessy Biosfera **3** (4), 91 (2004).
27. B. M. Vladimirskii, N. A. Temur'yants, and V. S. Martynyuk, *Space Weather and Our Life* (Vek 2, Fryazino, 2004).
28. A. N. Pavlov, *Impact of Electromagnetic Radiation on Life Activities* (Gelios ARV, Moscow, 2002).
29. S. E. Schnoll, V. A. Namiot, V. E. Zhvirblis, et al., Biofizika **29** (1), 153 (1983).
30. B. M. Vladimirskii and A. A. Konradov, Uch. Zap. Tavrich. Nats. Univ. im. V.I. Vernadskogo, Ser. Biol Khim. 20 (59), No. 1, 92 (2007).
31. T. A. Zenchenko, A. A. Medvedeva, N. I. Khorseva, et al., Geofiz. Protseccy Biosfera 12 (4), 73 (2013). www.biphys.ru/archive/rhythm00001.pdf
32. T. A. Zenchenko, M. Dzhordanova, L. V. Poskotinova, et al., Biophysics (Moscow) 59 (6), 1186 (2014). www.biophys.ru/archive/rhythm-00002.pdf

Translated by E. Yablonskaya