Optics and Spectroscopy, 2022, Vol. 130, No. 10

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Properties for the spectral parameters of intermolecular vibrations obtained using Raman scattering of light in dilute aqueous solutions of hydrogen peroxide and in the water

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Received August 06, 2021
Revised August 02, 2022
Accepted August 02, 2022

The spectra of Raman scattering (in the region $4-350\,\mathrm{cm}^{-1}$) of dilute aqueous solutions of hydrogen peroxide in a wide range of concentrations (from 3 to 0.024%), were measured. The spectral parameters of the high-frequency band of intermolecular (longitudinal) vibrations, obtained using the previously proposed approximating function, were analyzed. To do this, the spectral parameters of the band (for each experiment) were plotted on a diagram in which the abscissa axis is the square of the band width, and the ordinate axis is the square of the band frequency. Previously, it was shown that in such coordinates the water points are located in two regions separated by an inclined strip, in which spectral points (frequency gap) are practically absent. For points of hydrogen peroxide solutions with a concentration from 3 to 0.33%, all points are grouped near the straight line approximating them and are located below this frequency gap. For a series with extended range of peroxide concentrations from 3 to 0.024%, the scatter of points from the approximating straight line increases, but also most of the points are located below the gap. Of the rest of points, several points are near the edge of the gap, some of them are inside the gap, but the gap remains. It has been suggested that the presence of this gap is due to the fact that in the liquid phase some of the water molecules are bound to four neighbors, while the remaining molecules are bound to a smaller number of neighbors.

Keywords: Raman scattering, hydrogen peroxide, water, intermolecular vibrations, high-frequency band, inhomogeneous distribution, frequency gap, destruction of high-frequency states.

DOI: 10.21883/EOS.2022.10.54861.2621-22

1. Introduction

In this paper, we continue to study the properties of the spectral parameters of low-frequency (LF) spectra of Raman scattering (RS) for dilute aqueous solutions of hydrogen peroxide (HP) based on the ideas developed in the papers [1–8].

The spectra of intermolecular vibrations of water are determined by the properties and structure of hydrogen bonds between water molecules. The importance of the study of intermolecular vibrations for the study of liquids was realized almost immediately after the discovery of RS [9]. For more details on the structure of water and the history of the development of ideas of ideas about its structure, see [10,11], on the properties of bands of intermolecular vibrations of dilute aqueous solutions see in [2–8,10].

Note the following important points for understanding the features of the study of dilute aqueous solutions. In condensed phases, water molecules are bound by hydrogen bonding. Water molecule can have from one to four hydrogen bonds. In the works [11,12] it is considered that about half of the molecules are connected with 4 neighbors. Of the remaining ones, the vast majority is associated with 3 neighbors. Each of the classes forms a network that covers the entire space. These networks are nested within each other. These bonds are unstable in the liquid phase. They are constantly tearing and re-emerging. Their lifetime is on the order of picoseconds. The vibrations of these bonds are reflected in the IR absorption spectrum in the form of two broad bands with frequencies: high-frequency (HF) with a maximum of about $180\,\mathrm{cm}^{-1}$ and low-frequency (LF) with a maximum of about $50\,\mathrm{cm}^{-1}$. It is important for what follows, that there is some system with elastic bonds, the spectrum of which has two groups of lines forming two broad bands. The presence of other molecules in the solution leads to disruption of the grid of hydrogen networks of water molecules, which is reflected on in the observed spectrum.

In the literature, the shape of the RS bands in solutions, the positions of the maxima of these bands, and their widths under various conditions have been quite extensively studying. Based on the results of these studies, various aspects of the characteristics for liquids were elucidated. Among them are the inhomogeneity of the lines of intermolecular vibrations of water [13], the effect of acetone on the structure of water [14], the effect of temperature on the spectral parameters of water for various isotopes of oxygen and hydrogen [15]. As a rule, studies in solutions were performed mainly for high concentrations of dissolved substances (from 0 to several mol/l), when strong distortions in the spectrum of intermolecular vibrations, and these changes were used to draw conclusions about the structural structure of the liquid [14,15], were observed.

1.1. On the LF spectrum of intermolecular vibrations of water

The LF RS is located in the region $0-350\,\mathrm{cm}^{-1}$ and is quite well known [2,6]. The intensity of the optical spectrum $I(\nu)$ is a central maximum, monotonically decreasing to high frequencies, with some features in the region $100-200\,\mathrm{cm}^{-1}$ in the form of a low-intensity pedestal. This spectrum reflects both the properties of matter and the properties of scattering. To pass to the spectral characteristics of intermolecular vibrations of the solution, the optical spectrum $I(\nu)$ is transformed by multiplying by the reducing function, as a result, the reduced spectrum $\chi''(\nu)$ is obtained, which is called as the dynamic susceptibility (DS) [13,15]. It is associated with the density of states in the spectrum of vibrations.

In the DS spectrum, two clearly defined wide and partially overlapping bands are observed with lower frequencies of maxima than the IR absorption bands: about $45\,\mathrm{cm}^{-1}$ (LF band) and about $170\,\mathrm{cm}^{-1}$ (HF band). In [16–18] the interpretation of these two humps is given. At a maximum of about $170\,\mathrm{cm}^{-1}$ is the hydrogen bond stretching mode, at a maximum of $45\,\mathrm{cm}^{-1}$ is the bending mode of this bond, for which the break on the LF slope is associated with relaxation rotational motions of the molecule in the field of neighbors. It is conventionally to describe a spectral vibrational band in a form of Lorentz function with three spectral parameters: intensity, position of the maximum, and band width. In some papers, a more complex structure of these bands and other interpretations of the observed pattern are considered [6–8]. The explanation of the lower frequencies of the maxima of the bands at RS as compared with IR absorption given in Sec. 1 is described in our work [7] on the basis of the properties of a classical damped oscillator.

1.2. Function approximating the spectrum of intermolecular vibrations

To determine the spectral parameters of the bands, one used the approximating function for DS $\chi''(\nu)$ presented in [2,5–8], which has 9 fitting parameters. Of these, 6 refer to two Lorentzians describing 2 vibrational bands. These nine spectral parameters were determining by the least squares method by fitting the approximating function to the DS spectrum $\chi''(\nu)$ in the frequency range $4-320\,\mathrm{cm}^{-1}$. This function with the obtained parameters approximates well the spectrum [2,5–8]. It was found [3,5–8] that in addition to „regular“ changes in parameters, significant random changes in all spectral parameters are observed when the concentration of HP in water changes.

1.3. Display of the frequency parameters of the bands of the spectrum of intermolecular vibrations in frequency coordinates

As a rule, when studying the properties of solutions, the dependences (concentration, temperature) of each of the parameters were studied individually. In a series of the works [2–8,19,20] the LF of RS spectra for water and dilute aqueous solutions of HP were studied. In [3,5,6] the concentration dependences of all spectral parameters (each separately) were used to estimate the effective size (diameter) of the perturbation region of the hydrogen bond network near the HP molecule, which amounted to $6-7.5$ of the average distances between water molecules (taking into account the correction of these distances). Note that instead of concentration, we used the average distance between the molecules of the solute N_{av}, measured in average intermolecular distances of the solvent [3].

When analyzing the received materials, 3 important points were identified. First, all obtained DS spectra looked the same, with no visible differences. Secondly, all spectral parameters of both water and aqueous solutions of HP fluctuate. Third, the pairwise interdependences of the parameters showed that for both vibrations, the corresponding band widths and their positions of maxima are strongly correlated (hereinafter, we will call them as the frequencies or the observed frequencies and vibration band widths). Prior to the works [4–6], this issue was not considered in the literature. This is appeared in coordinates: observed bandwidth — observed band frequency [4–7]. Due to the properties of this dependence, which are explained below, it is more convenient to consider it in coordinates: width squared — frequency squared [7], which we call as frequency coordinates. In this coordinate system, each band is displayed as a point.

A few words of overview character about the results obtained earlier for water and HP solution to the formulation of the problem in this work. Detailed discussion of the problem is given below in the analysis of the results. In [7] the dynamic characteristics of the oscillators from the interdependence of the observed band frequencies and their widths are obtained. These are eigenfrequencies of oscillations and degrees of inhomogeneous broadening of the bands for water and HP. In this case, a small number of spectra (7 spectra) were recorded for water. For HP, solutions with a sufficiently high concentration of solutions ($0.3-3\%$) were measured. In this case, for both objects, the points for both water and HP were localized relatively close to the straight line approximating them. In [8] it was shown that a significant increase in the number of spectra under consideration (51 spectrum) leads to the fact that the region occupied by points in frequency coordinates, first, greatly increases, and second, the points are clearly divided into two sub-regions, separated by a fairly wide strip, in which no points practically presented ,i.e. they separated by a sort of frequency gap. In this regard, the question arises: if we expand the range of concentrations and increase the number of spectra under consideration, where will the new spectral points fall and how will they correlate with water points?

The purpose of this work is to study the effect of dissolved HP in a larger range of concentrations on the parameters of the high-frequency band of intermolecular vibrations of water and to study the spectral demonstration of this effect.

2. Recording of spectra

See [2,5,6] for details on recording of spectra. The spectra were recorded on a U-1000 spectrometer by Jobin–Yvon (France) in 90° geometry. The depolarized spectrum was recorded. The spectra were normalized to the maximum intensity of a very wide band of stretching vibrations located in the region $2500-4000\,\mathrm{cm}^{-1}$, which was used as an internal intensity standard.

3. Substances under investigation

Sealed ampoule with water for injection was used to record water spectra. To obtain of HP solution, the initial material was a medical 3% solution of HP. The solution was prepared immediately before recording. The concentration was controlled by the intensity of the characteristic line of HP $875.5\,\mathrm{cm}^{-1}$ for each recording. When approximating its shape, the Lorentz function [3,5,6] was used. All points of the dependence of the concentration measured from the intensity spectrum on the planned concentration during the preparation of the solution have fallen on proportional dependence with angular coefficient of 1.005 and a confidence interval of 0.007 [5]. Two series of HP solution spectra were recorded. The first series (series 1) contains 25 spectra recorded in a wide range of concentrations from 3 to 0.024%. In the second series (series 2), 17 spectra were recorded with a smaller concentration range: from 3 to 0.325%.

In the present work, one compare the spectral parameters of the HF band of the intermolecular spectra of HP solutions with the parameters of water. These spectral parameters were partly used in [7,8]. In [7,8] a small set of water spectra (7 spectra) is presented, in [8] a large set of water spectra (51 spectrum) is analyzed in detail.

4. Dilute solutions and vibration spectra

In terms of chemical composition and structural elements, the HP molecule is closest to the water molecule and also forms hydrogen bonds, however, the geometry of the HP molecule differs from the geometry of water molecules. Therefore, in an aqueous solution around the HP molecule in a certain region of finite size, the network of bonds of the nearest water molecules will be distorted, which should be reflected in the dynamic characteristics of water in the vicinity of the HP molecule (for example, the elasticity constants of bonds and their orientation, bond length, vibration frequencies of water molecules and the width bands) and, consequently, on the spectra of intermolecular vibrations of water. In the absence of solute associates at a very low concentration, these regions around each molecule will practically not intersect with each other, and the total volume of regions with a distorted bond network will be proportional to the number of dissolved molecules. As the concentration increases, the number of regions and their total volume will increase, and, finally, the undistorted structure will disappear. If the dynamic properties of water in the distorted regions are changed markedly, we can see changes in the spectrum. This makes it interesting to study the concentration properties of the spectral properties of dilute solutions

5. Results obtained for HP

Following [8], we also consider the properties of only the HF band of water oscillations.

Series 1 spectra of HP solutions (25 spectra) was recorded in a wide concentration range from 3 to 0.024%, which corresponds to intermolecular distances from 4 to 16.7. These data fluctuate strongly from spectrum to spectrum, so a series of 2 of 17 spectra was recorded for a smaller concentration range: from 3 to 0.325%, which corresponds to distances between HP molecules from 4 to 8.35.

It was shown in [4,5] that in coordinates of observed frequency — observed bandwidth, the points representing the spectrum are strongly correlated. In the figure, the circles show the points of series 2 of HP [4,5,7]. The values of the square of the observed Lorentzian width are plotted along the abscissa axis, and the squared frequency is plotted along the ordinate axis. The reasons for using the quadric values are explained below. Straight crosses denote data for a small set of water spectra (7 spectra) [7].

An explanation for this was given in [7] based on the concept of a classical oscillator, which is characterized by its eigenfrequency (EF) ν_0 and damping δ. It was also shown there that the observed maxima of the band in the DS [Bspectrum, its width Γ, and the EF are related by the formula

$$\nu_0^2 = \nu_m^2 + 0.5\Gamma^2/K^2. \tag{1}$$

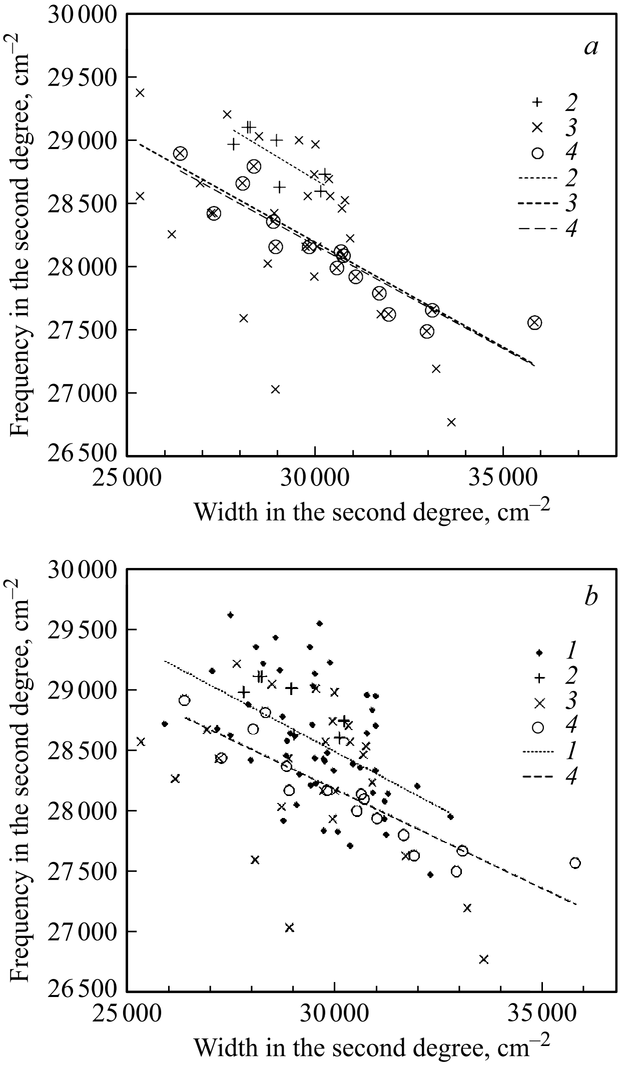
Here, the constant value K is the ratio of the observed width Γ to the homogeneous width γ determined by the oscillation damping. The quantity K is called as the inhomogeneous broadening coefficient (inhomogeneity coefficient). From the linear dependence (1), approximating the interdependence of the squares of the observed widths and positions of the band maxima, on the slope of this straight line, the inhomogeneity coefficient K is determined as

$$K^2 = -0.5(\partial\nu_m^2/\partial\Gamma^2)^{-1}. \tag{2}$$

For the small set of water spectra with the angular coefficient of -0.18, the inhomogeneity coefficient is 1.67. In (1) one can see that the free term of such a straight line is the square of EF. Whence the EF is 184.6 cm^{-1}, which is close to the known data on the IR absorption of water (183.4 cm^{-1} [7]). In our opinion, this is the main and most convincing confirmation of the correctness of the ideas developed in [7]. Knowing the coefficient K, one can obtain a homogeneous line width and, consequently, the decay time of vibrations [8]. It can be seen from (1) that the observed position of the spectrum maximum is always smaller than the EF vibrations and the shift is determined by the homogeneous width of the band of vibrations.

In [8] a substantially large set of spectra for water (51 spectrum) was considered in detail. The points in frequency coordinates occupy a much larger region there than the seven points considered above occupy. This can also be seen in the figure, *b*, where the points of the large set for water are indicated by blackened circles, and the small set for water (7 points) are indicated by straight crosses. The effective parameters of the set of 51 water points are [8]: the inhomogeneity coefficient $K = 1.65$ and the effective EF $\nu_0 = 184.3\,\mathrm{cm}^{-1}$, which is close to effective parameters of the set of 7 points given above. Nevertheless, in frequency coordinates (the square of the observed width —the square of the observed frequency) for the straight lines approximating both sets of water points at a squared width of 30000 cm^{-2} (near the center of the region occupied by the points), the difference between the squares of the frequencies is 205 cm^{-2}, i.e. they are noticeably separated.

In addition, the large set of water points [8] is divided, firstly, into two sub-regions, which are separated by a region containing practically no points, which can also be seen in the figure, *b*. Secondly, these two subdomains are in turn divided into several families of points approximated by straight lines, similar to those considered above [8], with their own approximating straight lines. Note that for HP solutions, when compared with the points of series 1, one of the points of series 2 can clearly be assigned to series 1. As a result, the straight line approximating the points of series 2 (figure, *a*, curve *4* — long dashes) has parameters slightly different from [7] . The angular coefficient for this straight line is $\alpha = -0.165$, the free term is $\nu_0^2 = 33107\,\mathrm{cm}^{-2}$, whence, by (3), the effective inhomogeneity coefficient $K = 1.74$ and effective EF $\nu_0 = 181.95\,\mathrm{cm}^{-1}$. For the entire complex of HP points (series 1 and 2 together: points with oblique crosses in the figure, *a*) the scatter of distances from the straight line approximating them (figure, *a*, curve *3* — short dashes) more than for series 2. In this case, this straight line practically merges with the approximating straight line of series 2 (figure, *a*). The parameters of the line for the entire complex of points of the HP: the angular coefficient of the line $\alpha = -0.167$, the free term $\nu_0^2 = 33188\,\mathrm{cm}^{-2}$. This gives $K = 1.73$ for the effective inhomogeneity coefficient, $\nu_0 = 182.18\,\mathrm{cm}^{-1}$ for the effective EF, which differs slightly from the series 2 parameters. In frequency coordinates for the straight line approximating the entire complex of HP points and the straight line approximating the points of series 2, with the same value of the squared width 30000 cm^{-2}, the difference between the frequency squares is 21 cm^{-2}, i.e.by an order of magnitude less than for two sets of water.



Dependences of the square of the observed frequency of a high-frequency oscillation on the square of its observed width: (○) — points of the series 2 for HP, are approximated by a dashed line *4* (long dashes), *a*: (□) — all HP points, approximated by lower dashed line *3* (medium size dashes), (+) — set of seven water points, approximated by dashed line *2* (short dashes); *b*: (+) — set of seven water points, (•) — 51 big set water point [8], which are approximated by a dashed line *1* (short dashes).

In the figure, *b* to compare solutions of HP and water, the above seven points for water (+), points of series 2 of HP (○) and series 1 (×) are superimposed 51 water points from data [8] (•). In [8] it was noted that for a large set of water points (51 points) in frequency coordinates, the empty region has the form of a strip with which are almost parallel sides, almost parallel to the straight line approximating the entire set of points (a kind of frequency gap). The lower edge of the empty region is close to this line and lies just above it. Above the empty region is almost 30% of all water points. In fact, this means that water, when recording these spectra at the same bandwidth, had a higher observed frequency of intermolecular vibrations. Based on [12], one can assume that this may correspond to states with a more rigid structure of the bond network, for example, with a larger average number of bonds per molecule due to an increase in the number of molecules associated with 4 neighbors. Note that the seven points of the small set of water presented above also lie near both boundaries of the empty region and actually form by themself the boundaries of the gap separating them. However, the points of this set, located near the upper boundary of the region free from the points of the large set, are noticeably distant from the upper boundary of this region. Thus, the empty region common to both sets was preserved, but narrowed. In [8] it was shown that the probability of distribution of points in frequency coordinates is non-uniform over the space of this coordinate system. Apparently, the boundaries of the empty region have the maximum probability of hitting points that correspond to the most probable water states. Therefore, in a set of 7 points, the spectra of more probable states were recorded, and all its points are located on both sides of the frequency gap near its boundaries. Moreover, this set itself consists of two sub-sets obtained at different times, and in each of these sub-sets the points were also located on both sides of the frequency gap near its boundaries.

In the figure, *b*, when comparing solutions of HP and water, a number of facts are clearly visible. First, only two points of series 2 are closely adjacent to the straight line approximating the entire set of 51 water points (empty circles *4*), all the rest are much lower, i.e., at an HP concentration of 0.33% or more, the states of water with HF are destroyed; it is possible that the average number of bonds of water molecules is reduced in these solutions. These concentrations correspond to average molecular distances between HP molecules of 8.35 the size of a water molecule and less.

Second, the observable empty region represented in [8] is preserved. Above the straight line approximating the entire set of 51 water points, there are 10 points of the series 1 of HP (oblique crosses *3*). The main part of the points of series 1 of HP did not fall into the frequency gap of the large sets of water, although six points of the series 1 are located directly near its upper and lower boundaries. Of these, several points also, although they fell inside the gap of a large set, are located near its boundaries approximately in the same places as the points of the water set of 7 points, so that the gap for the complex of all the points obtained has been preserved, although it has undergone some deformation. It is possible that the reason for falling points for the 7 spectra of water is the presence of a large amount of impurities in the samples during their recording.

Thus, we can assume that the probability that the points of the HP solution fall into the frequency gap in the distribution of water points (into the empty region) remains extremely low.

As a result of a comparison of the distribution of points of large sets of spectra of water and HP solutions in frequency coordinates, it can be unambiguously stated that the introduction of HP within the indicated limits reduces the observed frequencies of intermolecular vibrations of water in HP solutions.

6. Conclusion

It is shown that the approximating straight lines both for the points of series 2 of HP solutions with the absence of low concentrations ($< 0.35\%$) and for the entire complex of points for the HP solution with a large range of concentrations in frequency coordinates, the square of the observed width — the square of the observed frequency practically coincide, due to which practically coincide with their effective coefficients of inhomogeneity and effective EF practically coincide.

It is shown that the points representing the spectral parameters of the HF band of intermolecular vibrations of molecules in frequency coordinates for all the HP solutions studied by us do not fill the previously discovered empty region in the distribution of points representing the spectral parameters of pure water. The points representing the spectral parameters of the HF band for HP solutions with a concentration of more than 0.33% (the distance between HP molecules is 8.3 molecular distances and less) are almost all located below this empty region. Based on this, we can conclude that HP solutions with concentrations above 0.33% do not contain states with higher frequencies. Thus, we can conclude that the addition of HP reduces the observed frequencies of intermolecular vibrations of water in HP solutions.

Based on the existence of an empty region in the distribution of points for water in frequency coordinates, an assumption was previously made about a more rigid structure of the water network for states that are displayed as points above the empty region, due to an increase in the average number of bonds per molecule. The location of the points for HP then means that the presence of HP with a concentration of 0.33% or more reduces the average number of bonds of a water molecule in solution.

Conflict of interest

The authors declare that they have no conflict of interest.

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