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EFFECT OF SALINITY CONCENTRATION AND WATER TEMPERATURE ON THE RAYLEIGH WING ACCORDING TO FOUR-PHOTON POLARIZED SPECTROSCOPY DATA

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Abstract: Objective: The purpose of this study is to examine how the temperature of the water and the concentration of NaCl affect the Rayleigh wing line, with an emphasis on how supersonic lattice modulation contributes to the narrowing of the Rayleigh wing and the creation of maxima. **Method:** To examine the nonlinear optical characteristics of water and aqueous solutions, the study uses four-photon polarization spectroscopy under pre-threshold Brillouin scattering circumstances. The modification of nonlinear cubic susceptibility and its effect on anisotropic scattering were investigated under controlled experimental settings, such as varied salt concentrations and laser beam intensities. **Result:** The results show that higher water temperatures and NaCl concentrations improve adiabatic compressibility, which in turn raises the Brillouin scattering threshold and decreases the degree of nonlinear cubic susceptibility modulation. **Novelty:** This study provides new insights into the interplay between the anisotropy of Brillouin scattering, hypersound lattice modulation, and the structural changes in water induced by NaCl concentration and temperature variations, offering a deeper understanding of the microscopic mechanisms influencing water's scattering properties.

Keywords: Polarization, Anisotropy, Wing Lines, Brillouin Scattering, Quasicrystalline Lattice, Nac1, Rayleigh Wing Lines.

Introduction

It is known that in the framework of a mixed, two-fluid model of water, the existence of ice-like hydrogen-bound molecular complexes is assumed [1-3]. It is well established [4-5] that when registering the Raman spectrum of the OH band of water, a decrease in the averaging time or sampling interval up to the duration of one pulse (10ns) leads to significant fluctuations in the envelope of this band, manifested by an oscillation of its center by ±50 cm^{-1}. This coincidence indicates the existence of ice-like complexes in water at room temperature. Spectroscopy of small-angle scattering of X-ray synchrotron beams in the volume of water allowed us to identify components characteristic of massive ice [7-10].

In this paper, a model of Rayleigh line wing formation will be presented, taking into account the anisotropy of Brillouin scattering [6] on the scheme of four-photon polarization spectroscopy. Dilution of NaCl leads to disruption of the ice quasi-crystalline lattice formed by the H-bond in liquid water. Violation of the structure of quasicrystals should lead to a change in the nonlinear cubic susceptibility describing the process of stimulated Brillouin scattering, as well as anisotropic scattering.

Methods

This study employs a four-photon polarized spectroscopy technique to analyze the effects of salinity concentration and water temperature on Rayleigh wing formation in aqueous NaCl solutions. The experimental setup includes a laser source for generating the reference wave, a scattering medium for observing hypersonic lattice interactions, and a spectrometer for capturing the spectral distribution of the Rayleigh wing. The measurements were conducted across a range of NaCl concentrations (0.1–1.0 M) and temperatures (5–50°C) to determine their influence on anisotropic scattering and Brillouin thresholds.

Key parameters such as adiabatic compressibility, nonlinear cubic susceptibility, and Brillouin frequency shifts were evaluated under controlled environmental conditions. The study focused on detecting side maxima within the Rayleigh wing, corresponding to frequency shifts ranging from -2 cm^{-1} to 2 cm^{-1}. These shifts were analyzed for variations in amplitude and detectability, reflecting changes in the modulation degree of the anisotropic scattering process. Spectral data were processed using advanced software to ensure precise measurements of peak positions and amplitudes.

Control experiments were conducted to validate the findings, ensuring that observed spectral variations were attributable to salinity and temperature changes rather than instrumental artifacts. Additionally, comparative analyses were performed using deionized water as a baseline to isolate the effects of NaCl-induced lattice disruption. This comprehensive approach provides a robust framework for understanding how environmental factors influence Rayleigh wing narrowing and the optical properties of water systems.

Result and Discussion

The four-photon polarization spectroscopy of Rayleigh line wing allows one to obtain a new information about the structure and properties of liquids with strong interaction. In particular temperature and concentration deformations of the Rayleigh line wing narrow part $-2\ cm^{-1} \leq \omega_2 - \omega_1 \leq 2\ cm^{-1}$ were found in water and water solutions [1].

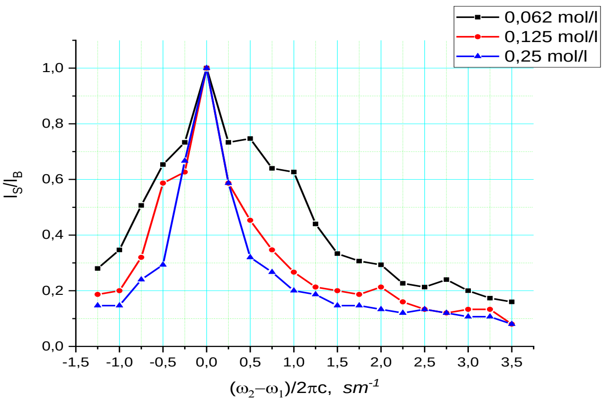

Figure 1. Spectra of four-photon polarization spectroscopy of the Rayleigh line wing in water at different salt concentrations

The results obtained showed (Fig. l) that salt concentration increase in water results in the Rayleigh line wing narrowing due to the contribution decrease into the Rayleigh line wing narrow part and the appearance of maxima shifted on Brillouin scattering frequency in Rayleigh wing line distribution (Fig. 2). The mechanism of the change of the Rayleigh line wing shape seems to be caused by the dynamical disturbances of ice like quasicrystal structure.

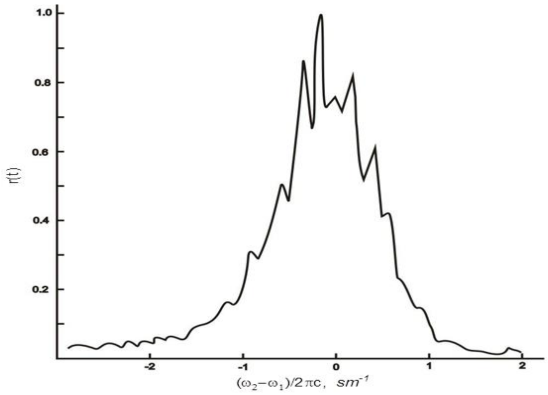

Figure 2. Rayleigh wing line distribution in pure water

The paper presents the Rayleigh line wing forming model taking into account the Brillouin scattering anisotropy [2] at four- photon polarization spectroscopy scheme. The dilution of NaCl results in the disturbance of ice like quasicrystal lattice formed by the H-bond in liquid water. The disturbance of quasicrystal structure is to lead to the change of nonlinear cubic susceptibility describing the process of stimulated Brillouin scattering $\left(\chi_{\mathrm{B}}^{(3)}\right)$ and anisotropic scattering $\left(\chi_{an}^{(3)}\right)$ as well.

It is known [2] that Brillouin spontaneous scattering is isotropic process as density fluctuations does not depolarized scattered radiation. However, in experiment were used laser radiation intensity close to the threshold values of Brillouin scattering. In a general case electrostriction is not isotropic process [3]. Even in isotropic dielectrics at a sufficiently great intensity of the electric field the electrostriction leads to the medium anisotropy. Meanwhile anisotropy nonlinear cubic susceptibility describing electrostriction is determined by the following way [3]:

$$\Delta\chi^{k}(\omega) = \frac{1}{4\pi}\frac{\partial\varepsilon}{\partial\rho}\Delta\rho \qquad (1)$$

where

$$\Delta p = (\frac{\gamma}{2\pi v^2})|E^2|,\ \gamma = \rho\frac{\partial\varepsilon}{\partial\rho}$$

electrostriction coefficient, v-hypersound velocity; E- tenseness of the electric field. Thus the hypersound lattice is not only the lattice of density fluctuation, but anisotropic fluctuation as well.

It is known [1] the resonance of nonlinear cubic susceptibility χ_B- can be observed only at $\omega_2 - \omega_1 = \Omega$ (where Ω - Brillouin resonance frequency). Then for the four-photon Rayleigh line wing scattering intensity one may write:

$$I_s = \left|\frac{\chi_{an}}{i \pm (\omega_2 - \omega_1 \pm \Omega)/\delta\nu_{an}}\right| I_1^2 I_2^2 L \qquad (2)$$

here I_1 and I_2 - the intensities of laser beams with frequencies $\omega 1$ and ω_2 accordingly, L - nonlinear interaction length, $\delta\nu_{an} = 10cm^{-1}$ - the halfwidth of anisotropy scattering line at water temperature 24 °C [6].

While conducting the experiments strict control is observed of the absence of the Brillouin scattering at the moment of the pumping radiation interaction with the medium in question, that is the investigation are carried out in the pre-threshold regime of Brillouin scattering. However one can may state that in this case there exists a hypersound lattice in the medium, though Brillouin scattering is not registered by device.

Using the dependence for the intensity P of the hypersound lattice on li and Brillouin scattering intensity IB as well:

$$P = \left|\frac{G I_1}{2\alpha}\right| \left|\frac{\Omega}{\omega_1}\right| I_B \qquad (3)$$

where ***a*** - hypersound absorption coefficient, G - Brillouin scattering amplification increment it can be shown that with the decrease of the Ii by 20 % the Brillouin intensity decreases by 400 times, i.e. Brillouin scattering can be registered in the active spectroscopy scheme [1], which is used in the experiment given. That is why in the scheme of the experiment it is necessary to take into account the anisotropy of the hypersound lattice, which is caused not only by the $(\omega_2 - \omega_1)$frequency, but by the pumping beam (ω_1) interaction with the medium as well.

For the anisotropic scattering intensity the expression can be written:

$$I_S = \left|\frac{\chi_{an}^{(3)}}{i + (\omega_2 - \omega_1)/2\delta\nu_{an}}\right|^2 I_1^2 I_2^2 L \qquad (4)$$

The hypersound generation leads to the modulation of nonlinear cubic susceptibility of the medium accordingly (1):

$$\Delta\chi^{(3)} = \frac{1}{4\pi}\frac{\partial\varepsilon}{\partial\rho}\left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)\frac{1}{2\pi v^2}|E|^2 \approx const|E|^2 \qquad (5)$$

i.e. the modulation of $\chi^{(3)}$ will take place due to the change of E. When (ω_1) interact with the medium the wave $2E\cos\Omega t$ is formed. This wave causes the hypersound waves. Changeable radiation $(\omega = \omega_2 - \omega_1)$interact with $2E\cos\Omega t$ and this interaction leads to the modulation of the cubic nonlinear susceptibility of the medium:

$$\chi^{(3)} = const\left(2\cos\Omega t + \cos\omega t\right) \qquad (6)$$

or for the intensity of Rayleigh line wing:

$$I_s = \left|\frac{const*(2\cos(\Omega t) + \cos(\omega t))}{i + \frac{\omega}{2\delta\nu_{an}}}\right|^2 I_1^2 I_2^2 L. \qquad (7)$$

As it is known the phase difference of two waves has a great importance when they interact and so the expression (7) will have a form:

$$I_s = \left|\frac{const*(2\cos(\Omega t) + \cos(\omega t + \Delta\varphi))}{i + \frac{\omega}{2\delta\nu_{an}}}\right|^2 I_1^2 I_2^2 L. \qquad (8)$$

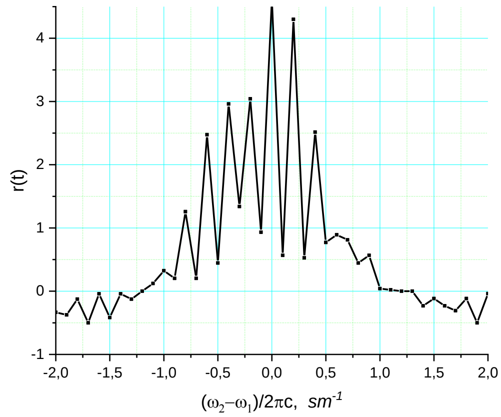

Figure 3. Distribution of Rayleigh wing lines taking into account the anisotropy of the hypersonic lattice at low concentrations of salt C1

The obtained dependence shows that the modulation of nonlinear cubic susceptibility by Hypersound lattice does lead to the appearance of the maxima shifted to Brillouin frequency in Rayleigh line wing, that is observed in the experiment (Fig. 1,2).

Obtained intensity distribution of the rayleigh line wing is symmetric with reference to the zero frequency. Fig. 3 and fig. 4 show that the change of leads to the violation of maxima distribution symmetry relative to zero frequency and also a change of the relative height, which is testified in the experiment (Fig. 1,2).

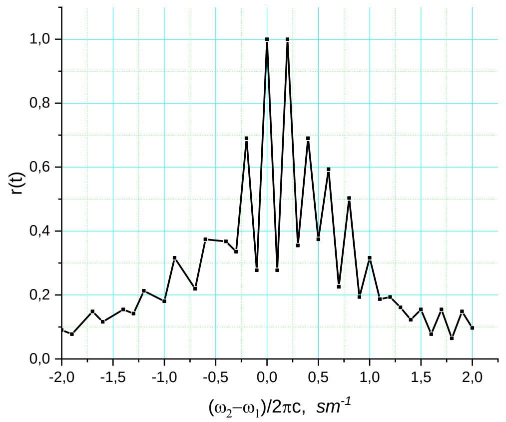

Figure 4. Distribution of Rayleigh wing lines taking into account the anisotropy of the hypersonic lattice with an increase in the concentration of salt C2 (C2 > C1)

As it is known the threshold intensity and affectivity of Brillouin scattering depend on active medium. Results obtained in [6] shows that the threshold in NaC1 solutions (C=3%) have more high values then in pure water and scattering affectivity decrease essentially. We explain such dependence by the gydrotation causes the adiabatic compressibility decrease.

It is known that water temperature increase leads to the adiabatic compressibility decrease. That is why only those maxima it will be registered which intensity is high then registration threshold of the device.

NaC1 concentration and temperature increase in water causes the Brillouin scattering

Threshold and the narrowing of Rayleigh wing line.

Conclusion

Thus it was demonstrated that the modulation of nonlinear cubic susceptibility by hypersound lattice makes it possible to explain the appearance of maxima in Rayleigh line wing intensity distribution, observed in the experiment [1] and NaCl concentration and temperature increase in water leads to the narrowing of Rayleigh wing line.

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