Hypersound harmonic generation by means of stimulated Brillouin scattering.

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Introduction

The interference effects between stimulated Brillouin, four-photon scattering and hypersound harmonic generation can be a promising and probably powerful method for detection of low frequency features of Rayleigh wing spectra in liquids. Particularly, in liquid water such approach can gives the information on temperature derivatives of the Kerr constant and orientational relaxation time, which strongly depends on the short-range order in liquid water and the type of intermolecular interactions. The coherence of the four-photon mixing also provides interference of molecular and clusters resonance contributing to the spectrum of scattering, which enables one to enhance or suppress a given resonance by appropriately choosing the experimental conditions.

Four-photon spectroscopy is based the process $\varpi_\Delta = \varpi_p - (\varpi_p - \varpi_S)$ [1-3], where the difference $\varpi_p - \varpi_S$ is scanned in the vicinity of studied resonances. In our case it is tuning around the Brillouin doublet and the Rayleigh wing. A measured parameter is the polarization of the ϖ_Δ wave, determined by a nonlinear source, which is proportional to the third-order susceptibility of the medium.

Four-photon spectroscopy uses the high intensity of laser beams. Therefore, to describe the signal, one has to consider not only the interaction of pumping and scattering waves but also the specific amplification of signal and reference waves in the pump field, owing to the stimulated scattering of the pump wave and hypersonic harmonics generation. Furthermore, in the studied low-frequency spectroscopic band, when a frequency difference $\varpi_p - \varpi_S$ is less than 10 cm^{-1}, the strongest mechanisms of optical nonlinearity are strictional and orientational interactions (Kerr effect). In the present paper we experimentally observed the hypersound harmonic generation with the help of four-photon spectroscopy in Rayleigh wing range of spectrum. The theoretical model of this interaction is presented also.

Experiment

A scheme of the experimental setup is shown in Fig. 1. The radiation of the master oscillator *(1)* (a single-frequency Nd:YAG laser with a pulse duration of 10 ns and a repetition rate of 5 Hz) is split into two channels and amplified in a one- or two-stage amplifier *(2)*. The second harmonic is generated in a thermally stabilized CDA crystal *(3)*. The radiation of the third harmonic generated in the DKDP crystal *(4)* is used to pump a Coumarin 500 (C-500 from Exciton) dye laser *(5)*, with the wavelength tunable between 485 and 565 nm and a bandwidth of less than 0.1 cm^{-1}. Glan-Thompson polarizers *(6)* are used to sharpen the linear polarization of pumping beams. Circularly polarized light is obtained using a quarter-wave plate *(7)*.

Two counterpropagating waves with electric field intensities $\boldsymbol{E}_S$ and $\boldsymbol{E}_p$ and frequencies ω_S and ω_p generate a four-photon signal in a thermally stabilized cuvette *(8)* containing liquid water. The four-photon signal is detected with a PMT *(10)*. The waves with $\boldsymbol{E_S}$ and $\boldsymbol{E_p}$ are linearly and circularly polarized, respectively. The nonlinear wave-mixing gives rise to a wave with frequency $\omega_\Delta = \omega_S$ and intensity $I_\Delta \propto \left|\chi^{(3)}\right|^2 I_p^2 I_S$, where I_Δ, I_p, I_S is the field intensity and $\chi^{(3)}$ is the third-order susceptibility. The polarization vectors of wave $\boldsymbol{E_\Delta}$ resulting from four-wave mixing and the pumping wave $\boldsymbol{E_S}$ are noncollinear and can be separated by a polarization prism *(6)*.

Figure 2 shows the four-photon spectra of double distilled water H_2O and heavy water D_2O, which were received with a spectral resolution 0.15 cm^{-1}. One can see that both spectra show the Brillouin resonances with spectral separation 0.55 and 0.7 cm^{-1} correspondingly and some peaks with 1.13 and 1.57 cm^{-1} spectral separation. These peaks spectral position corresponds to second harmonics of Brillou hypersound waves.

Discussion.
Nonlinear Phase Matching for Resonance Scattering at Double Brillouin Frequency .

Experimentally observed resonance has no conventional explanation when the signal frequency is equal to $\omega_\Delta = \omega_p + 2\Omega$, where Ω corresponds to the Brillouin hypersound frequency

$$\Omega \approx 2(v/c)\cdot\omega_P ,$$

here v is the sound velocity. Wave vector of the microwave lattice created by interaction of the signal (ω_Δ) and pumping (ω_p) optical beams is equal to $\boldsymbol{q} = \mathbf{k}_\Delta - \mathbf{k}_p$ ($|q| = k_\Delta + k_p$). However we can not indicate any resonance modes in the spectrum of our media which corresponds to such combination of parameters. Moreover, as we mentioned, this kind of resonance scattering was observed when the pumping power was high enough for observation of high intensity stimulated Brillouin scattering in the direction collinear to the direction of the pump wave.

Thus the Stokes and the anti-Stokes components of acoustical and optical waves are able to take part in phase matching of resonance scattering when $\omega_\Delta = \omega_p + 2\Omega$. Below we propose the mechanism of nonlinear phase matching for media over the threshold of stimulated Brillouin scattering. The mechanism is based on electrostriction effect and four photon interaction due to Kerr effect responsible for background Rayleigh wing scattering. We take into account interaction of anti-Stokes optical wave with the signal beam and parametric coupling of the Stokes and anti-Stokes hypersound waves. The least is provided by an electrostriction pressure P_{ES}:

$$P_{ES} = \frac{1}{8\pi}\left(\frac{\partial\varepsilon}{\partial\rho}\right)\rho\cdot E^2$$

where ε is the dielectric permeability, $\boldsymbol{E}$ is the electric field strength, $\rho = \rho_0 + \rho_\approx$, ρ_0 is the equilibrium density of medium and $\rho_\approx$ is hypersound perturbation of the density. In the frame of mechanism under consideration an intermediate electromagnetic field $\boldsymbol{E}$ is generated due to four phonon interaction with the signal $\boldsymbol{E_\Delta}$ and anti-Stokes $\boldsymbol{E_A}$ waves. Interaction of this wave with the signal and the Stokes hypersound wave (ρ_S) creates the acoustic wave ρ_A at the Brillouin frequency Ω, which direction of propagation corresponds to the anti-Stokes acoustic wave.

This wave plays a part of the dynamic lattice for scattering of the pumping beam (ω_p) and for generation of an optical wave $\boldsymbol{E_A}$ at the anti-Stokes frequency $\omega_A = \omega_P + \Omega$. Such mechanism of the resonance scattering for signal frequency $\omega_\Delta = \omega_p + 2\Omega$ can be described by the system of coupled equations:

$$\frac{\partial E'}{\partial z} = \gamma_R E_A^2 E_\Delta^*$$

$$\rho'_A = \frac{1}{D_R}\left(\rho_0\frac{\partial\varepsilon}{\partial\rho}\right)\frac{\rho_S^*}{\rho_0}\left(\frac{E_\Delta E'^*}{8\pi\rho_0 v^2}\right)$$

$$D_R = \frac{(2\omega_\Delta - 2\omega_A - \Omega)^2}{\Omega^2} - 1 + iQ_\Omega^{-1}$$

$$\omega_A = \omega_P + \Omega$$

$$\frac{\partial E'_A}{\partial z} = -\frac{i}{2}k\left(\rho_0\frac{\partial\varepsilon}{\partial\rho}\right)\frac{\rho'_A}{\rho_0}E_P$$

where Υ_R is the amplitude of four photon interaction, Q_Ω is the quality factor of hypersound modes at frequency Ω. The resonance feature of scattering is reflected by the factor D_R equal to i/Q_Ω when $\omega_\Delta = \omega_p + 2\Omega$. The scattering efficiency defined by the ratio of amplitudes of the scattered and signal waves in the output of medium is equal to:

$$\left|\frac{E'_A}{E_\Delta}\right| = \frac{1}{6}\left(\frac{\Gamma_R}{\Gamma_B}\right)\left|\left(\frac{E_A^{\max}E_\Delta}{E_P^2}\right)\left(\frac{\rho_S^{\max}}{\rho_0}\right)\right|\frac{1}{|D_R|Q_\Omega}$$

where Γ_R and Γ_B are the increments of the four photon and Brillouin scattering respectively, E_A^{max} and ρ_S^{max} correspond to maximal values of this variables in the output of the interaction area. At the resonance condition $|D_R|Q_\Omega = 1$ in a strongly nonequilibrium supercritical state when $|E_A^{max} E_\Delta / E_P^2| \sim 0.5$ the efficiency can be estimate as $|E'_A/E_\Delta| \sim 10^{-3}$-$10^{-4}$ that is in agreement with the experimental data as well as the sensitivity of the efficiency to the signal wave intensity.

Conclusions

The performed experiment and theory confirm the specific interference mechanism between the stimulated Brillouin scattering, hypersonic and pump waves, that forms the fine structure of four-photon scattering spectrum in the Rayleigh wing spectral range. This mechanism can be used in four-wave experiments for measurement of some parameters, which are hardly measurable in other schemes. In particular, it concerns the temperature derivatives of the Kerr constant and orientational relaxation time, which strongly depend on the short-range order in liquid water.

References

1. Y.R.Shen *The Principles of Nonlinear Optics;* Wiley-Interscience: New-York, 1984.
2. A.F.Bunkin, G.A.Lyakhov, .A.Nurmatov, N.V.Suyazov. Appl.Phys. B 66, 91-94, 1998.
3. A.F.Bunkin, G.A.Lyakhov. Physics of Vibrations, v.8, 21-31, 2000.

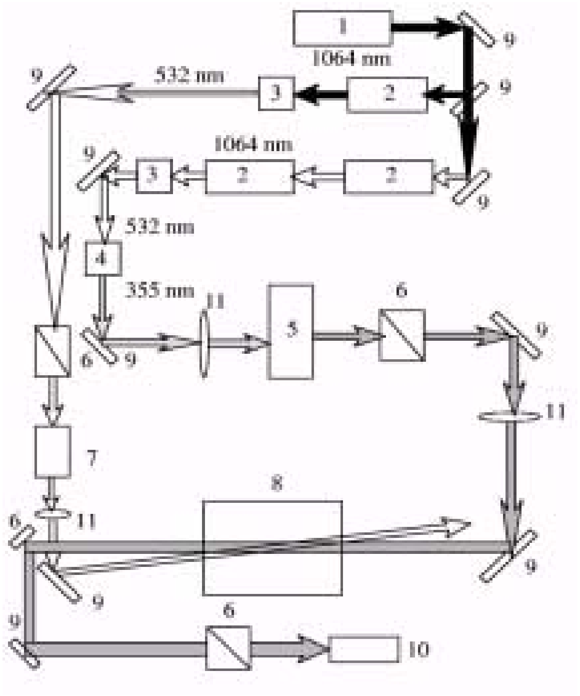

Fig. 1. Scheme of the experimental setup: (*1*) Nd:YAG laser, (2) Nd:YAG amplifiers; *(3)* CDA crystals for second harmonic generation; *(4)* DKDP crystal for third harmonic generation; *(5)* Coumarin 500 dye laser; (6) Glan-Thompson polarizers; (7) quarter-wave plate; *(8)* sample; (9) mirrors; *(10)* photodetector; *(11)* focusing lens.

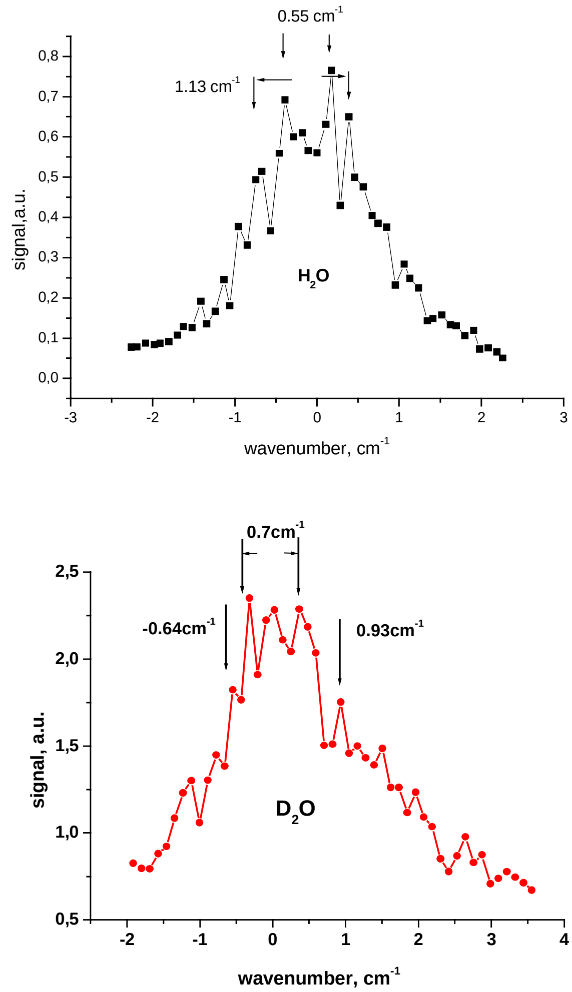

Fig.2 The four-photon spectra of double distilled water and heavy water.