

Influence of convection on the stimulated concentration light scattering

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Abstract. A non-linear growth of the scattering intensity and the frequency shift of the spectral lines of scattered light close to the half-width of the spontaneous scattering in the back scattering of light in the suspensions of latex nanoparticles in water were found. It proves that we observed a stimulated scattering of light on the particle concentration variations. Influence of convection is taken into account using Doppler measurements of fluid flow.

1. Introduction

Each type of spontaneous light scattering has a corresponding type of stimulated scattering (SS). SS attributes are a non-linear increase in the scattered light intensity under increasing of input pump laser radiation, and frequency shift in the spectral line of scattered light, or change of the shift for SBS, STS. In case of absence of frequency shift of the spontaneous scattering line, the value of the SS line shift, in accordance with a steady state model, should be equal to half width of a spectral line of the corresponding spontaneous scattering. An interference grating of intensity is created by interference of the exciting and back scattered waves. It results in the refractive index grating, at which SS occurs [1].

The attempts to study stimulated concentration light scattering (SCoLS) - analogue of spontaneous concentration light scattering (SpCoLS) on concentration fluctuations were undertaken repeatedly in gas mixtures [2-4], in aqueous solutions [5], in the gold nanotubes suspensions [6-8].

Application of correlation spectroscopy and of continuous laser allowed us to observe the frequency shift of SCoLS on particles in liquid in contrast to previous works [6-8].

Frequency shift (the cosine component of the correlation function) and a non-linear increase in intensity SCoLS were measured first time in suspensions of silicon nanoparticles in oil, diamond in water [9] and latex nanoparticles in water [10]. It was found that the cosine component amplitude and the ratio of the scattering intensity I_{sca} to the power of the input laser beam P_{las} increased with laser power P_{las} increasing i.e. I_{sca} depends on P_{las} as a square or even more sharply.

All these facts give reason to believe that we are dealing with stimulated light scattering on variations of the particles concentration in liquid, i.e. SCoLS, analogous with the complex phenomena occurring when a STLS or SSRLW take place [11].

2. Application of dynamic light scattering to study of stimulated light scattering

In the process of stimulated concentration light scattering the interference of exciting and scattered waves creates the interference grating of intensity. Maxima (or minima) of the grating draw the



particles of suspension in [12, 13]. It results in creation of the grating of particle concentration and refractive index.

$$N(z,t) = N_0 + \delta N(t) \exp(-i\Omega t + i|\vec{k}_L - \vec{k}_S|z) \quad (1)$$

where $N(z,t)$, N_0 are local and mean particle concentration, $\delta N(t)$ - the amplitude of concentration grating, z - coordinate, \vec{k}_L and \vec{k}_S - wave vectors of the exciting and scattered light.

Due to the half-width of the unshifted line of light spontaneously scattered by particles in the fluid is Dq^2 , where D - diffusion coefficient, and $q = |\vec{k}_s - \vec{k}_L|$ - scattering wave vector, the SS process should lead to the appearance in the scattered light spectrum of the lines shifted relative to the excitation light to the same value $\Omega_{st} = Dq^2$, similar to STS, where $\Omega_{STS} = \chi q^2$.

$$W_{st} = Dq^2 = \frac{k_B T}{6\rho h R} \left(\frac{4\rho n}{I_{532}} \sin\left(\frac{q}{2}\right) \right)^2 = \frac{k_B T}{6\rho h R} \left(\frac{4\rho n}{I_{532}} \right)^2 \quad (2)$$

where k_B - Boltzmann's constant, T - temperature, η - viscosity of the medium, R - radius of the particles, n - refractive index of the medium, λ_{532} - wavelength of green laser.

Note that the grating of concentration (1) moves with the velocity $V_{gr} = \Omega_{st}/q$.

For particle of 480 nm radius in water the theoretical calculation of SS shift from (2) gives values of $478 \pm 44 \text{ sec}^{-1}$ or $76 \pm 7 \text{ Hz}$. The shift of such a quantity can be observed by correlation spectroscopy and can not be detected in experiments with setups using Fabry-Perot interferometers.

If the scattered light reaching the square photodetector has two lines, which are shifted relative to each other on the value of Ω , then the cosine component will appear in the correlation function of the scattered light due to the frequency beats. Analogous to [14], we can derive an expression for the intensity autocorrelation function $G^{(I)}(\tau)$ (ICF) of the scattering intensity in case of beats between the patch of the exciting light (I_h) and a line of stimulated scattering (I_{st}) in the presence of the spontaneous scattering (I_{sp}) [10]:

$$G^{(I)}(t) = I_{sp}^2 \exp(-2G_{sp} t) + I_{st}^2 \exp(-2G_{st} t) + 2I_{sp} I_h \exp(-G_{sp} t) + 2I_{st} I_h \exp(-G_{st} t) \cos(Wt) + A \langle I \rangle^2 \quad (3)$$

where Γ_{sp} is $G^{(I)}(\tau)$ the half-width of the spontaneous scattering line, Γ_{st} - half-width of the stimulated scattering line. In the case of plane waves and steady state $\Omega = \Omega_{st} = \Gamma_{sp} = Dq^2$. In the case of a stimulated process, the intensity of shifted line increase exponentially with laser pump increase, and at low intensities it should be square-law.

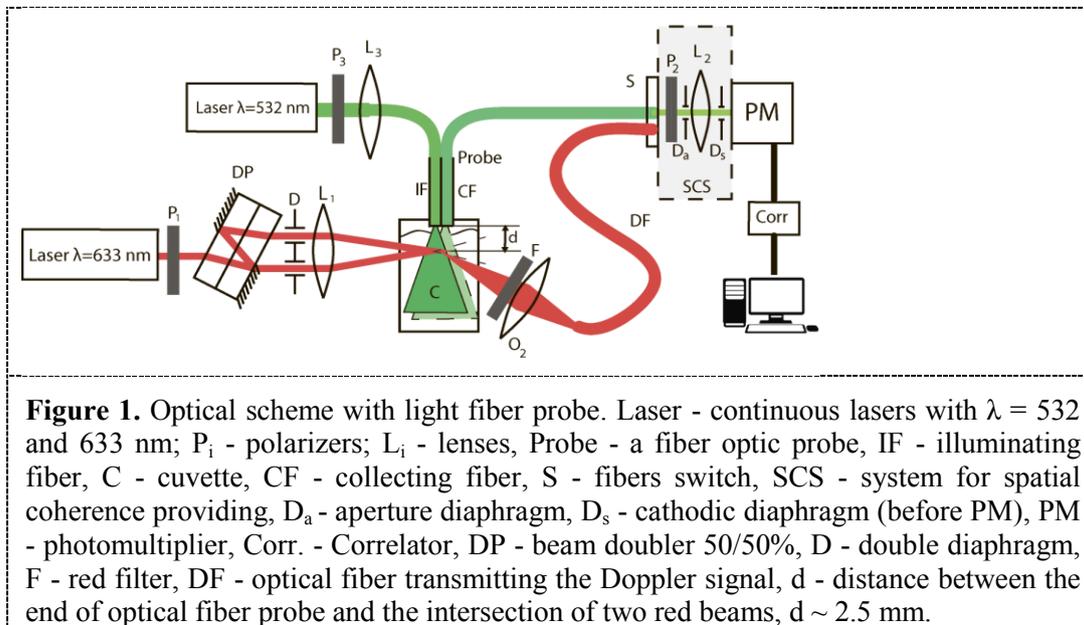
3. Samples and experimental setup

For samples of latex preparation we used monodisperse suspension of spherical latex particles of radius $r = 480 \text{ nm}$ in water produced by company FSUE "NIISK" with a narrow size distribution and with an initial concentration of 6% by weight. We diluted the suspension by dust-free double-distilled water to a volume concentration of polystyrene $C_v = 7 \times 10^{-4}\%$.

Fig. 1. shows a scheme for registration of SCoLS. The light source was a continuous solid state laser with a wavelength $\lambda = 532 \text{ nm}$.

The laser beam, having passed through polarizer P_3 , which allows us to adjust the power of the exciting radiation, was focused by lens (Lens, $f=15 \text{ mm}$) on the illuminating fiber IF input. The end of optical fiber probe includes the output of the illumination fiber IF and input of the collecting fiber CF. These ones are parallel and placed in direct vicinity to one another. The axes of the optical fibers are at a distance 0.3 mm. The probe immersed in the cuvette containing the suspension. Collecting fiber CF of probe collected light from the scattering volume. The scattered light from the collecting fiber, passing through the polarizer P_2 and a system for spatial coherence providing, SCS, arrived to the cathode of a photomultiplier. The PM pulses were sent to a correlator for obtaining SS ICF. SCS

system consisted of aperture D_a , cathode diaphragms D_s , and a lens O. Light fiber system required almost no adjustment, except of the input of illuminating light to optical fiber and alignment of the collecting fiber end image to the diaphragm D_s . It was necessary only to make sure that the light from the fiber probe don't fall on the cell wall.



To determine and measure the role of convection velocity, we introduce to the optical circuit He-Ne laser beam, vertical doubler DP and the focus lens L_1 ($f=10$ sm), composing typical two-beam Doppler circuit, as described in [15]. The light scattered at a small angle relative to the axis of the He-Ne laser in a horizontal plane focused by the lens O_2 through a red filter F into the fiber DF comes in SCS, to the cathode of a photomultiplier PM, and PM counts passes to correlator for ICF accumulation. It was easy to move from measurement of SS to vertical velocity measurement by switching of fibers CF and DF at the entrance of the SCS system.

Cosine period of ICF of scattered light of two He-Ne laser beams gives us convective flow velocities V_c :

$$V_c = \frac{W_c / I_{532}}{4pn} = \frac{I_{633}}{2T_{633} \sin(\alpha/2)} \quad (4)$$

where Λ is a period of Bragg grating, λ_{633} – the wavelength of He-Ne laser, T_{633} - cosine period of ICF obtained through the fiber DF, α - the angle of convergence of beams in the air.

The velocity of convective flow is measured at the distance of about 2.5 mm from the end of the fiber probe. This distance is estimated to be the most effective for SS registration from distance dependence of the overlapping area of the cones of illumination region of IF and CF vision field and from the dependence $I_{sp} \sim 1/r^2$.

4. Results and discussion

Fig. 2. shows an increase in the amplitude of the cosinusoidal component of the correlation function with an increase of pump power for latex nanoparticles suspension with radius $r = 480$ nm., and Fig. 3, 4. shows the scattered light intensity increasing with pump power increasing for the particles. These two facts indicate the presence of SS.

In the experiment we have simultaneously measured total SS spectral shifts Ω_{exp} under convection influence and velocities of convection flow V_c when the fiber probe was directed up and down. The

values of the convection velocity V_c was calculated from the period of the cosinusoidal ICF component T_{633} of light collected through the optical fiber DF, according to (3). From the value of the

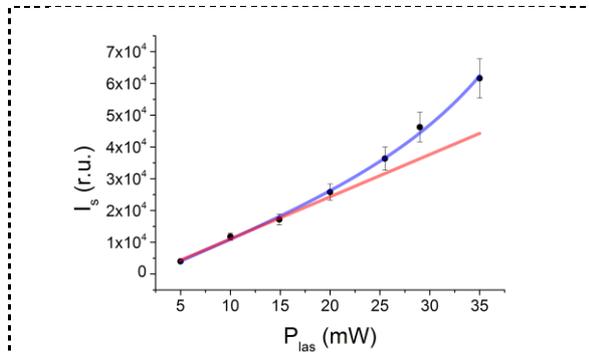
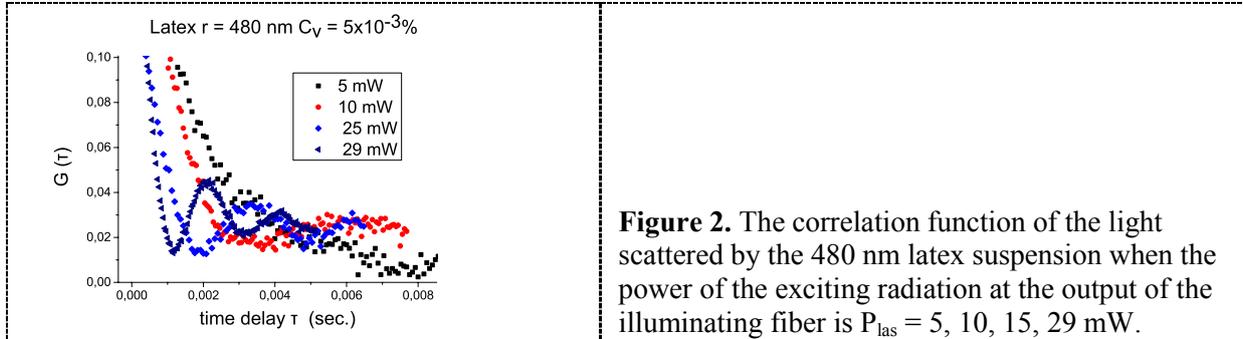


Figure 3. Dependence of the intensity of scattering in the latex suspension on the pump power P_{las}

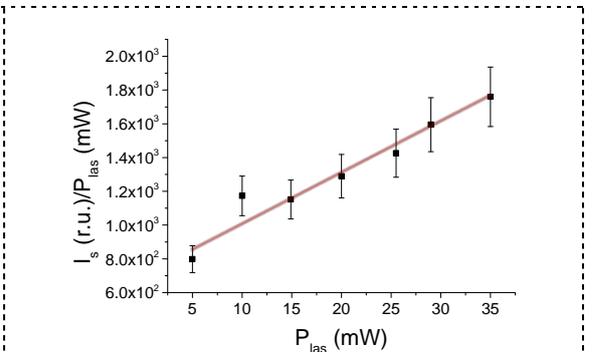


Figure 4. The ratio of the scattering intensity and the pump power P_{las} on the pump power P_{las} for the latex suspension [11].

period of ICF cosinusoidal component T_{532} of light collected through the optical fiber CF, we calculated the total velocity of the concentration SS grating V_{exp} :

$$V_{exp} = \frac{W_{st}/I_{532}}{4\rho n} = \frac{I_{532}}{2T_{532}n} \quad (5)$$

Fig. 5,6. represents the dependences of the velocity of convective flow V_c and the total concentration SS grating velocity V_{exp} on the power of the exciting beam. One can see that for the position of the fiber probe “down” $V_{exp} < V_c$. This indicates that in such position of fiber the speed of the wave concentration V_{gr} relative to the fluid is directed from the fiber and opposite to the velocity of convection flow V_c is more then V_{exp} , and $V_{exp} = V_c - V_{gr}^{DOWN}$ indicating that SS frequency shift is Stokes. For the position “up” $V_{exp} > V_c$, therefore $V_{exp} = V_c + V_{gr}^{UP}$, velocities V_c and V_{gr} are directed in the same direction and have be added, so as shift SS is also Stokes. This shift is defined as:

$$W_{exp} = \frac{4\rho n V_{gr}}{I_{532}} \quad (6)$$

Fig. 7 shows clean SS spectral shift, calculated from V_{gr} by (5). For the pump power more than 15 mW (when there is SS) the mean SS frequency shift is $\Omega_{st(exp)} = 643 \pm 55 \text{ s}^{-1}$. The discrepancy between this experimental value and theoretical one $\Omega_{st} = 478 \pm 44 \text{ s}^{-1}$ is of about 12%.

5. Conclusion

So we believe that we have detected presence of SS on the particle concentration variation in the liquid and measured the frequency shift of the spectral line of the SS, earlier [11] in a steady state regime, and now in condition of the convection presence.

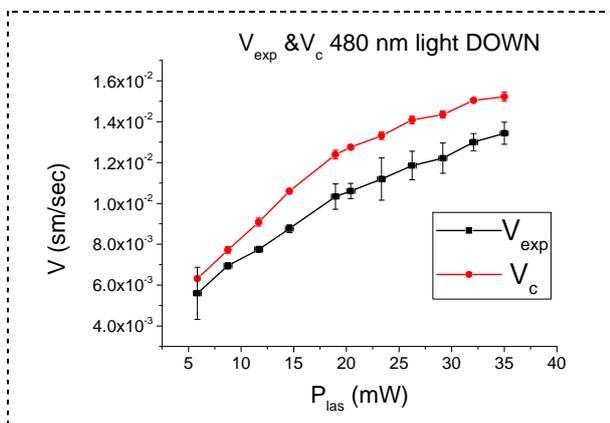


Figure 5. The dependence of the total velocity of the concentration SS grating V_{exp} and convection flow velocity V_c on the power of the exciting beam at the position of the light fiber probe down

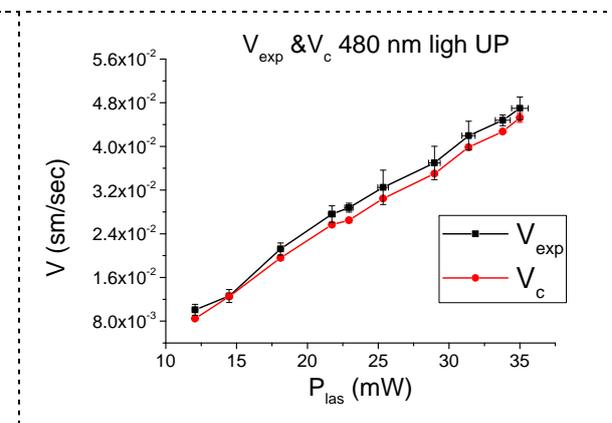


Figure 6. The dependence of the total velocity of the concentration SS grating V_{exp} and convection flow velocity V_c on the power of the exciting beam at the position of the light fiber probe up

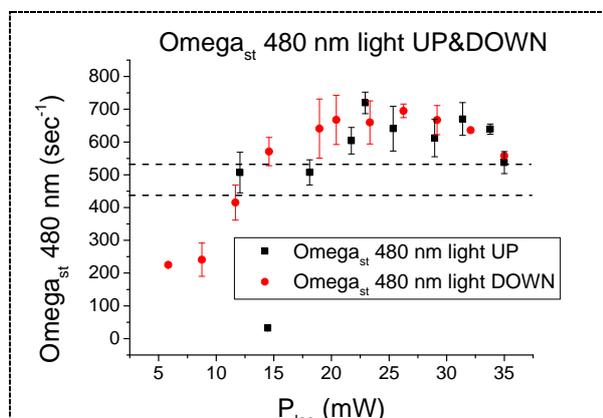


Figure 7. The values of the SS spectral shift obtained from formula (5) under increasing in the pump power. Theoretical limits are denoted by shaded lines.

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