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RAYLEIGH SCATTERING IN METAL NANOPARTICLES IN LIQUIDS

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Abstract

In this study, silver nanoparticles with narrow particle size distribution synthesized by acoustoplasma technique were studied. A number of features of stimulated thermal Rayleigh scattering (STRS) in pure liquids and suspensions of nanoparticles were investigated in this work. It is shown that the scattering efficiency is not reduced in case of wide spectral bandwidth pump radiation. It is shown experimentally that the frequency shift of the scattered signal relative to the pump frequency greatly exceeds the theoretical value. It is also shown theoretically that the frequency shift value does not depend on the linewidth of the pump.

Keywords

Nanoparticles – Silver – Light – Scattering

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Introduction

Recently, some works were published devoted to the stimulated scattering of laser radiation in suspensions of nanoparticles¹. It is supposed that stimulated Mie scattering is observed². The results were obtained with different suspensions of nanoparticles, both metallic and dielectric in different liquids (water, toluene, hexane, chloroform e.g.)³. A main characteristic of this type of scattering is the low pump threshold intensity (usually below than threshold of stimulated Brillouin scattering) and the absence of frequency shift of the reflected radiation relative to the frequency of the pumping⁴.

A necessary condition for the observation is the presence of a linear or two-photon absorption of the pump radiation by nanoparticles⁵. Authors of above cited works supposed that the mechanism of stimulated thermal scattering in this case is absent⁶.

They believe that in stimulated scattering process take place a convolution of pump laser spectral line with amplification line of Rayleigh scattering⁷. In addition, in case of convolution the scattering efficiency should decrease at broadband pumping radiation because the amplification coefficient is actually inversely proportional to the line width of the pumping radiation that is not observed experimentally⁸.

In this work, we theoretically and experimentally shown that in the case of pure liquids, and so at the use of nanoparticle suspensions occurs STRS associated with the absorption (linear or two-photon)⁹.

¹ N. A. Bulychev; M. A. Kazaryan; L. S. Lepnev; A. S. Averyushkin; M. N. Kirichenko; A. R. Zakharyan y A. A. Chernov, "Luminescent properties of nanoparticles synthesized in electric discharge in liquid under ultrasonic cavitation", Proceedings of SPIE, Vol: 10614 num 13 (2018).

² Yu. V. Ioni; S. V. Tkachev; N. A. Bulychev y S. P. Gubin, "Preparation of Finely Dispersed Nanographite", Inorganic Materials, Vol: 47 num 6, (2011): 597-602.

³ M. N. Kirichenko; N. A. Bulychev; L. L. Chaikov; M. A. Kazaryan y A. V. Masalov, "Effect of iron oxide nanoparticles on the concentration-versus-sizes relation of proteins in the blood plasma and serum, and in model solutions", Proceedings of SPIE, Vol: 10614 num OM (2018).

⁴ Yu. P. Aleksandrova; N. S. Budanova; A. A. Farmakovskaya; N. S. Okorokova; G. N. Ustyuzhaninova; N. P. Zharova y V. Kohlert, "Organic pigments surface modification by isobutyl vinyl ether copolymers under the action of ultrasonic", Revista Inclusiones, Vol: 7, num Especial (2020): 11-21.

⁵ N. A. Bulychev; E. L. Kuznetsova; V. V. Bodryshev y L. N. Rabinskiy, "Nanotechnological Aspects of Temperature-Dependent Decomposition of Polymer Solutions", Nanoscience and Technology: An International Journal, Vol: 9 num 2 (2018): 91-97.

⁶ N. A. Bulychev; M. A. Kazaryan; E. S. Gridneva; E. N. Murav'ev; V. F. Solinov; K. K. Koshelev; O. K. Kosheleva; V. I. Sachkov y S. G. Chen, "Plasma discharge with bulk glow in the liquid phase exposed to ultrasound", Bulletin of the Lebedev Physical Institute, Vol: 39 num 7 (2012): 214-220.

⁷ M. N. Kirichenko; L. L. Chaikov; I. S. Burkhanov; N. A. Bulychev y M. A. Kazaryan, "Effect of the pH of iron oxide nanoparticles solution on the rate of fibrin gel formation (according to light scattering data)", Proceedings of SPIE, Vol: 11322 num 1E (2019).

⁸ N. A. Bulychev; M. A. Kazaryan; A. Ethiraj y L. L. Chaikov, "Plasma Discharge in Liquid Phase Media under Ultrasonic Cavitation as a Technique for Synthesizing Gaseous Hydrogen", Bulletin of the Lebedev Physical Institute, Vol: 45 num 9 (2018): 263-266.

⁹ A. V. Ivanov; V. N. Nikiforov; S. V. Shevchenko; V. Yu. Timoshenko; V. V. Pryadun; N. A. Bulychev; A. B. Bychenko y M. A. Kazaryan, "Properties of Metal Oxide Nanoparticles Prepared by Plasma Discharge in Water with Ultrasonic Cavitation", International Journal of Nanotechnology, Vol: 14 num 7/8 (2017): 618-626.

Light scattering studies of suspensions of nanoparticles

The experiments were carried out with the second harmonic of a neodymium laser $\lambda=0.53 \mu\text{m}$. As "solvents" was selected toluene and hexane¹⁰. Note that even pure toluene has an appreciable two-photon absorption, and pure hexane did not. In figure 1, absorption spectrum of investigated substances in region of two-photon resonance of our pumping radiation is designated by arrow¹¹. As nanoparticles, silver nanoparticles were used, which were obtained in two different ways (laser ablation and plasma synthesis)¹². For the theoretical explanation as well as examination of such materials it is referred to¹³.

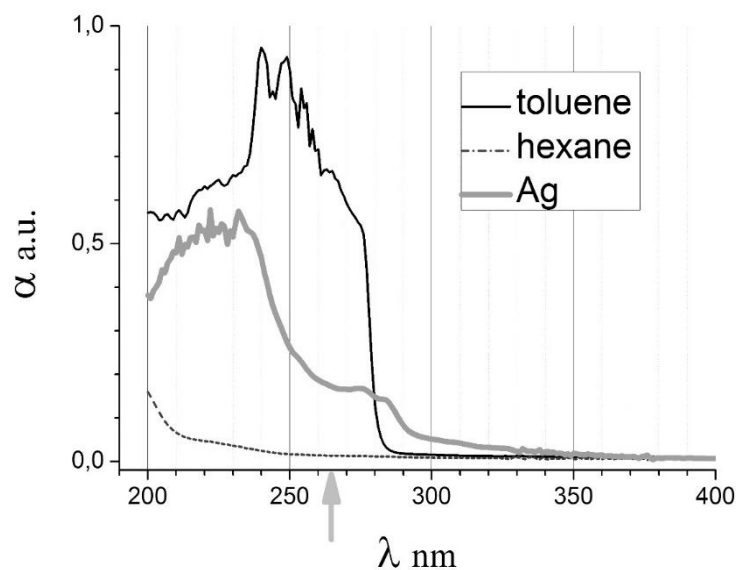


Figure 1

Absorption spectra of liquids and silver nanoparticles suspension

Fabry-Perot interferometer with a dispersion range $6.94 \cdot 10^{-2} \text{ cm}^{-1}$ was used to measure the spectral shifts. In pure hexane due to the absence of two-photon absorption thermal Rayleigh scattering was not observed¹⁴. On the contrary, in pure toluene, the

¹⁰ N. Bulychev; W. Van Camp; B. Dervaux; Y. Kirilina; K. Dirnberger; T. Schauer; V. Zubov; F. E. Du Prez y C. D. Eisenbach, "Comparative Study of the Solid-Liquid Interface Behaviour of Amphiphilic Block and Block-like Copolymers", *Macromolecular Chemistry and Physics*, Vol: 210 (2009): 287-298.

¹¹ K. V. Pushkin; S. D. Sevruk; N. S. Okorokova y A. A. Farmakovskaya, "The most efficient corrosion inhibitors for aluminum anode of electrochemical cell used as a controlled hydrogen generator", *Periodico Tchê Quimica*, Vol: 15 num 1 (2018): 414-425.

¹² Yu. P. Aleksandrova; N. S. Budanova; A. A. Farmakovskaya; N. S. Okorokova; G. N. Ustyuzhaninova; N. P. Zharova y V. Kohlert, "Ultrasonic treatment impact on the stability of aqueous dispersions of inorganic and organic pigments in the presence of surfactants", *Revista Inclusiones*, Vol: 7, num Especial (2020): 387-397.

¹³ Yu. O. Kirilina; I. V. Bakeeva; N. A. Bulychev y V. P. Zubov, "Organic-inorganic hybrid hydrogels based on linear poly(N-vinylpyrrolidone) and products of hydrolytic polycondensation of tetramethoxysilane", *Polymer Science Series B*, Vol: 51 num 3-4 (2009): 135.

¹⁴ I. S. Burkhanov; L. L. Chaikov; N. A. Bulychev; M. A. Kazaryan y V. I. Krasovskii, "Nanoscale metal oxide particles produced in the plasma discharge in the liquid phase upon exposure to ultrasonic cavitation. 2. Sizes and stability. Dynamic light scattering study", *Bulletin of the Lebedev Physical Institute*, Vol:41 num 10 (2014): 297-304.

stimulated temperature Rayleigh scattering was observed at the level of pumping ~ 3 mJ. Fig. 2 (left) shows the interferogram of the pump radiation and the stimulated Rayleigh scattering¹⁵. The lower half of the figure corresponds to the pump radiation. Top – line stimulated Rayleigh scattering.

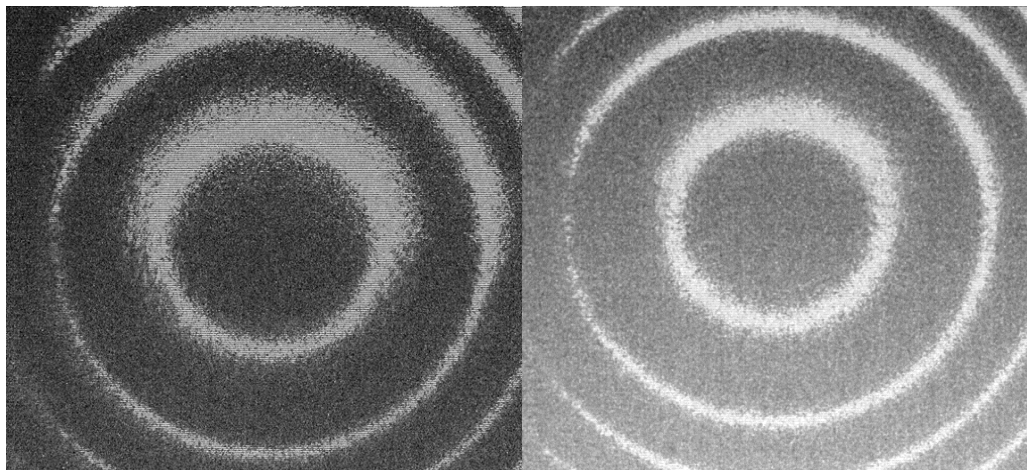


Figure 2

Interferogram of the pump radiation and the stimulated Rayleigh scattering (left) and interferogram of radiation back scattered in toluene with nanoparticles (right)

In the result of the interferogram processing, it was obtained that the Rayleigh scattering line is frequency shifted by 160 MHz relatively to the line of the pump into anti-Stokes region¹⁶. This value significantly exceeds anti-Stokes shift, which is predicted by theory¹⁷. This difference may indicate that the microscopic value of the thermal diffusivity can be much larger at small ~ 170 nanometers range, the characteristic period of the thermal grating¹⁸. Figure 2 (right) shows the interferogram of radiation back scattered in toluene at a concentration of nanoparticles ~ 1 gramm/liter. It is clearly seen that in addition to anti-Stokes shifted components we have a relatively wide anti-Stokes wing¹⁹.

Processing of the interferogram showed that the anti-Stokes shift stays the same ~ 150 MHz, and the anti-Stokes wing has a value ~ 300 MHz. Similar results were obtained in hexane with the same concentration of nanoparticles.

¹⁵ V. N. Nikiforov; N. A. Bulychev y V. V. Rzhetskii, "Elastic properties of HTSC ceramics", Bulletin of the Lebedev Physical Institute, Vol: 43 num 2 (2016): 74-79.

¹⁶ Yu. P. Aleksandrova; N. S. Budanova; A. A. Farmakovskaya; N. S. Okorokova; G. N. Ustyuzhaninova; N. P. Zharova y V. Kohlert, "Theoretical and experimental studies of the spectral characteristics of doped semiconductors using zinc oxide and sulfide", Revista Inclusiones, Vol: 7, num 3 (2020): 453-463.

¹⁷ A. V. Rudnev; N. G. Vanifatova; T. G. Dzherayan; E. V. Lazareva y N. A. Bulychev, "Study of stability and dispersion composition of calcium hydroxyapatite in aqueous suspensions by capillary zone electrophoresis", Russian Journal of Analytical Chemistry, Vol: 68 num 8 (2013): 700.

¹⁸ N. A. Bulychev; M. A. Kazaryan; A. S. Averyushkin; M. N. Kirichenko; A. R. Zakharyan y A. A. Chernov, "Dynamic characteristics of electric discharge in liquid under ultrasonic cavitation", Proceedings of SPIE, Vol. 10614 num 14 (2018).

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Conclusions

From our experimental results it follows that the addition into pure toluene nanoparticles does not change qualitatively the picture of the stimulated thermal scattering, though the presence in fluid of nanoparticles leads to the appearance of additional absorption mechanism. Our main result is the observation of anomalously large anti-Stokes shift. This allows us to draw definitive conclusions about the thermal relaxation mechanisms of the refractive index gratings.

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