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Temperature oscillations during photoinduced heating of aqueous suspensions of silicon nanoparticles

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Abstract. Temperature oscillations (pulsations) were detected in aqueous suspensions of silicon (Si) nanoparticles NPs under laser irradiation with highly absorbed light. The temperature pulsation frequency was found to depend on the NPs concentration in suspension and laser irradiation power. The observed phenomenon is assumed to be caused by the local overheating of Si NPs close to the boiling point of water, while the average heating of the surrounding liquid was insignificant. The observed phenomenon is discussed in view of potential applications in local photo-induced hyperthermia of cancer.

1. Introduction

In recent years, nanoparticles (NPs) of various substances and materials have been widely studied as an alternative to traditional methods of cancer treatment [1]. It was found that NPs do not enter most of the tissues, they cannot go beyond the walls of healthy blood vessels. However, tumor tissues have increased vascular permeability, and nanoparticles can penetrate them. Therefore, the use of NPs in the diagnosis and treatment of oncological diseases is very promising, because NPs can provide an improvement in the patient's quality of life without causing strong negative effects. Moreover, pure silicon (Si) NPs should not be toxic to the body. Therefore, this work considers Si NPs, which are not only low-toxic but also biodegradable and they can also be used in various methods of hyperthermia [2,3].

The essence of the photohyperthermia method is that optical radiation absorbed by tissues is converted into heat, which, even at a sufficiently high temperature, destroys cancer cells. It is known that silicon nanoparticles with sizes from 10 to 100 nm, which absorb light in the visible and near-infrared ranges of the spectrum well enough, can accumulate in the tumor region near cell membranes and penetrate cells. It was found that the energy of optical radiation absorbed by NPs is converted into heat, while cancer cells in the area of localization of NPs undergo thermal destruction. The widespread introduction of the photohyperthermia method into medical practice is also facilitated by using reliable, inexpensive, and compact emitters based on powerful single laser diodes in the spectral range of 800 - 808 nm in the transparency window of biological tissues. For such emitters, quasi-continuous and



pulsed generation modes have been developed, and the problems of ensuring reliability, design simplicity, and low cost of the required power and cooling drivers have been solved [3].

However, with photohyperthermia in continuous irradiation, there is a risk of damage to healthy tissues surrounding the tumor, limiting this method's use. To increase the efficiency and safety of the photohyperthermia method, it seems promising to use a repetitively pulsed irradiation regime, which prevents a significant increase in the average tissue temperature [4].

In this paper, we found quasi-periodic pulsations that appear during pulsed laser-induced heating of aqueous suspensions of Si NPs. However, to understand the results obtained in this work, a theoretical analysis of the heating of silicon nanoparticles in aqueous suspensions is required. But at the same time, the observed effect, in our opinion, is promising for use of Si NPs for cancer therapy applications [6-8].

2. Experimental part

Photo heating was performed using a solid-state nanosecond Laser Compact - TECH 351 laser (see Fig.1), which included a generator providing a pulse frequency of up to 5 kHz; during the experiments, the duration was 20-50 ns, and the average laser power was 70-75 mW. The wavelength of laser radiation was selected using light filters; the following wavelengths were used in the experiments: 351 nm, 526 nm, 1053 nm.

Thermographic measurements were carried out using a FLIR C3 thermal imager and a Seek Thermal Scanner.

For the first part of our experiments, we used Si NPs (ACS Material Advanced Chemical Supplier) obtained by HF decomposition of silane. As can be seen from the data of scanning electron microscopy (SEM) in Fig. 2, NPs had an average size of 15 nm and an almost spherical shape.

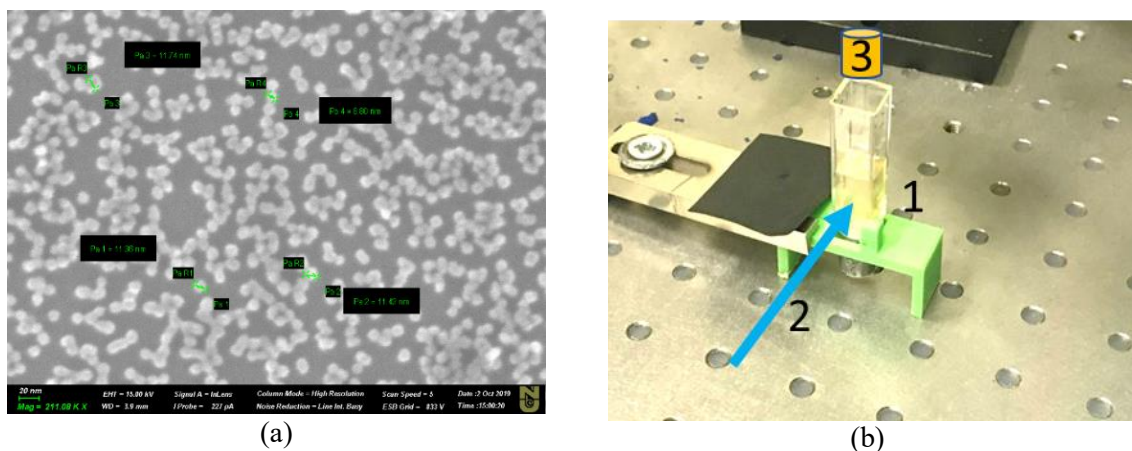


Figure 1. (a) SEM image of Si NPs; (b) experimental set-up, where 1 is a cuvette with Si NPs suspension, 2 is a laser beam, 3 is a thermal imager.

Aqueous suspension of sSi NPs was placed in a cuvette at a distance of 30 cm from the laser. The NP concentrations from 0.25 to 1 mg/ml were used.

3. Results and discussion

3.1 Manifestation of quasiperiodic pulsations in aqueous suspensions of nanoparticles.

During experimental studies, it was found that upon photoexcitation of an aqueous suspension of silicon NPs with an average size of 15 nm, along with a general increase in temperature, its pulsations take place in a certain region. Fragments of video recording are shown in Figure 2. Figure 2 shows a frame-

by-frame fragment of a video recording of thermographic control for 1 mg/ml concentration, radiation power 70 mW, and frequency 1 kHz.

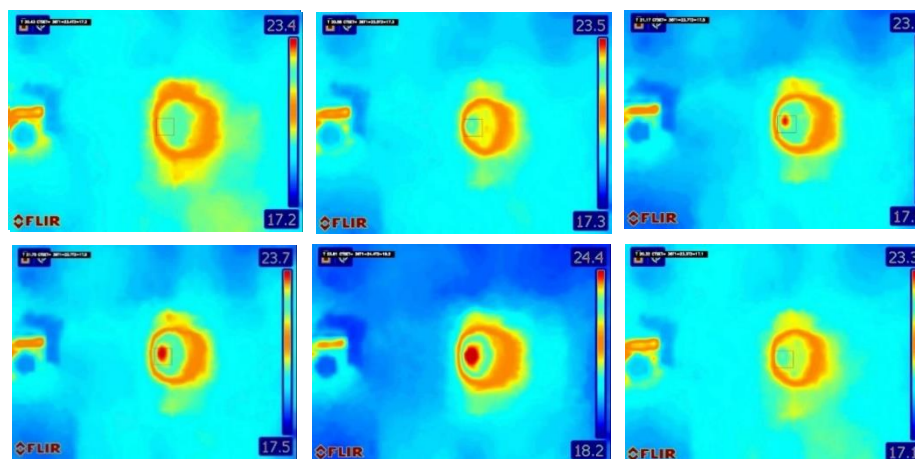


Figure 2. Frames of a video fragment of the thermographic control of photoheating of Si NPs with an average size of 15 nm and a concentration of 1 mg / ml, irradiation power 70 mW, frequency 1 kHz.

3.2 Study of the dependence of the frequency of manifestation of quasiperiodic pulsations in suspensions of nanoparticles.

To study the dependence of the frequency of occurrence of pulsations, the following studies were carried out, the laser power was changed at a constant concentration of the suspension, and the concentration of the suspension was changed at a constant laser power. The time dependence of the appearance of pulsations was also investigated. To estimate the ripple frequency, the records were processed using the software developed by our team. Figure 3 shows graphs of temperature changes in the process of photoexcitation of a suspension with a concentration of 1 mg/mL with a reference power decrease from 70 mW to 35 mW.

The experiments carried out on the photoheating of aqueous suspensions of silicon nanoparticles showed that the size of the thermal focus remains unchanged during the process of photoexcitation, from which it can be concluded that it is possible to localize the heating focus inside an oncological neoplasm.

In the process of performing experimental studies, pulsations of temperature pulsations were found. It can be assumed that the effect of pulsations is associated with large absorption of light at 351 nm, which allows accumulating a large amount of energy in a small volume, which leads to intense vaporization (boiling) near NPs at the initial stage with the subsequent unification of boiling fronts. Vaporization, in turn, generates the transfer of hot liquid from the epicenter to the periphery, which leads to cooling and a repetition of the cycle, also as an indirect confirmation of the hypothesis.

In the second part of our work, 10-150 nm sized Si NPs prepared by femtosecond laser ablation of crystalline Si targets in water [4] were investigated. The prepared Si NPs were selected by size and the Mie resonance for which corresponds to 630 nm. In the process of photoheating these nanoparticles in an aqueous suspension with a concentration of 0.2 mg/ml using a continuous laser with a wavelength of 633 nm, power of 30 mW, quasiperiodic oscillations were also observed (see Fig. 4).

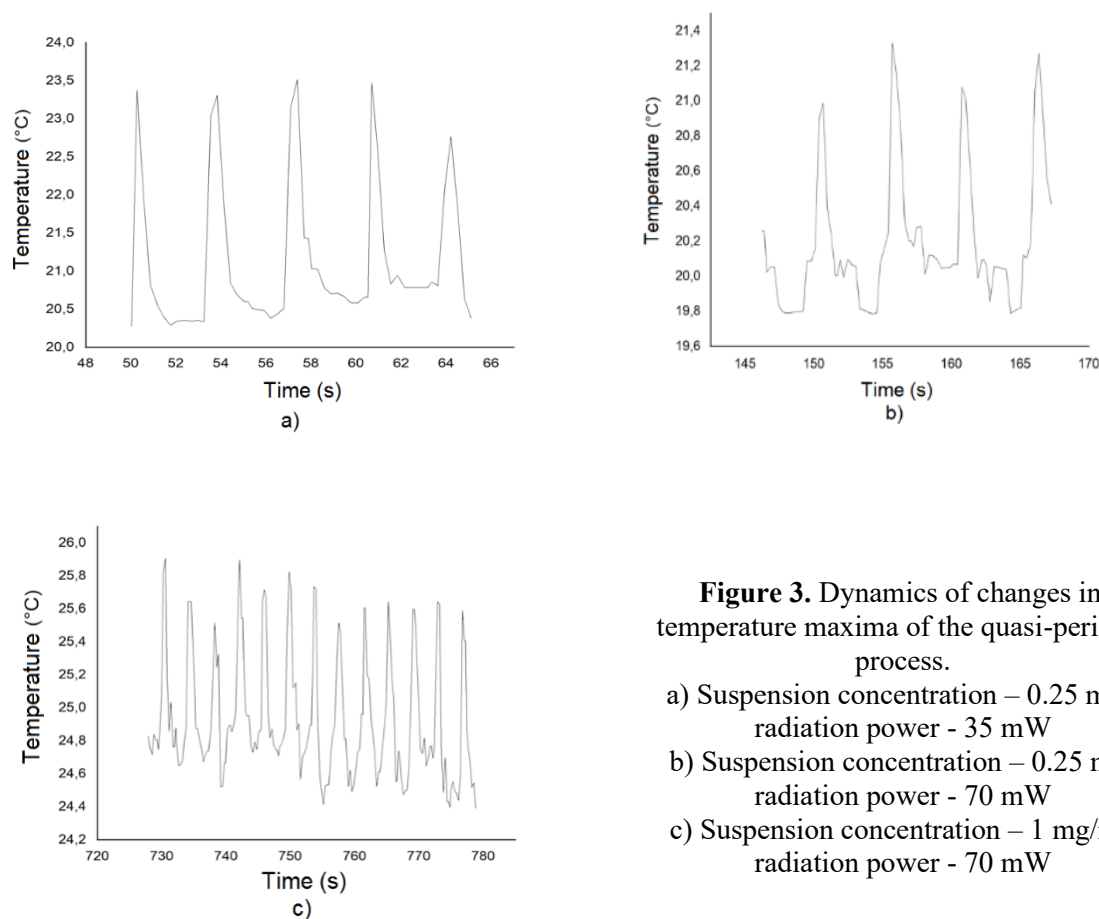


Figure 3. Dynamics of changes in the temperature maxima of the quasi-periodic process.

- a) Suspension concentration – 0.25 mg/ml, radiation power - 35 mW
- b) Suspension concentration – 0.25 mg/ml, radiation power - 70 mW
- c) Suspension concentration – 1 mg/ml, radiation power - 70 mW

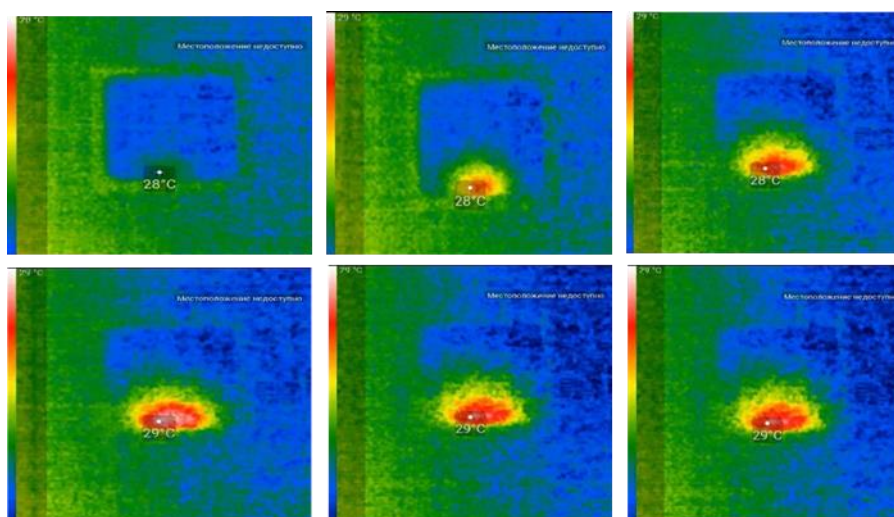


Figure 4. Frames of a video fragment of the thermographic control of photoheating of Si NPs with the average size of about 100 nm and a concentration of 0.2 mg/ml, irradiation power 30 mW, wavelength 633 nm.

Based on the above hypothesis about the occurrence of oscillations, the manifestation of oscillations in resonant particles indicates a promising possibility of using Si NPS in biomedicine.

4. Conclusions

Quasiperiodic change in the temperature of aqueous suspensions of Si NPs was observed when they were irradiated with highly absorbed laser radiation. It is assumed that the indicated phenomenon is caused by the local heating of NPs to the boiling point of water, or close to it with insignificant average heating of the surrounding liquid. Analysis of the data obtained after studying the found effect suggests that a significant effect on the pulsation frequency largely depends on the frequency of laser pulses, their power, and the concentration of Si NPs in the suspension. The observed phenomenon can be used in the method of local hyperthermia of cancer because overheated Si NPs will act as localizing non-toxic photothermal agents.

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