Photonic nanojets generated by alumina microstructures with different surface morphology

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Abstract. The interaction of optical radiation with dielectric microparticles, leading to the emergence of field spatial localization called the “photonic nanojet” was numerically and experimentally studied within the wide range of environment refractive indexes. It is established that at a certain ratio of the microparticle and the environment refractive indexes, as well as the curvature of the particle profile, the photonic nanojet effect can be used to locally excite the sensitive layer of fluorescent sensors of chemical compounds in liquid and gaseous media.

1. Introduction
The term “photonic nanojet” (PNJ) was first used in [1], where the process of optical radiation interaction with dielectric microparticles was demonstrated by numerical methods. At present, the PNJ is applicable in such areas as super resolution microscopy, Raman and fluorescence spectroscopy. To achieve high resolution it is necessary to ensure the highest radiation intensity along with a minimum half-width of the localization region of PNJ [2-4]. At the same time, the possibility of using the PNJ in the field of optical sensorics is considered rarely. For instance, the effect of luminescence enhancement was discussed in [5–7], but it was only noted about the perspective to use the PNJ to register various kinds of chemical and biological compounds, and the studies were not carried out. In addition, most theoretical studies of the PNJ are carried out for arbitrary values of the refractive index of the medium \( n_c \), such as, air \( (n_c = 1) \), water \( (n_c = 1.33) \) and the “average” refractive index of chemosensitive polymers \( (n_c = 1.5) \) [1–4]. Meanwhile, the refractive index of the polymers can vary over a wide range from 1.545 to 1.4 when measured in dry and swollen form, respectively [8, 9].

In this regard, the aim of the work is numerical simulation and further experimental study of the PNJ parameters generated by dielectric microstructures with different surface profiles based on aluminum oxide depending on the dispersing media refractive index.

2. Materials and methods
Numerical calculations of the geometrical parameters of aluminum oxide microstructures and the PNJ generated by them, were carried out using specialized software by the finite difference time domain method. The cell size of the spatial grid was at least 1/50 of the wavelength of the exciting radiation \( \lambda = 532 \text{ nm} \).

The dielectric microstructures of various morphology dispersed in a layer of a chemosensitive polymer were formed by the method successfully tested earlier for TiO\(_2\) [10]. In the first stage, ordered
arrays of holes in the layer of the chemosensitive chitosan polymer were obtained on quartz substrates by lithography. Next, microspheres of aluminum oxide with a diameter of D = 2 μm (Corpuscular Inc, USA), dispersed in a 1% solution of polymethyl methacrylate (PMMA), were deposited to the cellular structure by spin-coating. Microstructures of a given “crescent” and “cone” morphologies were obtained by directed etching of aluminum oxide microspheres in hydrofluoric acid. For this purpose, a layer of 5% PMMA solution was additionally deposited to cover a part of the surface of the microspheres. In the case of “crescent” particles, 1/4D of microspheres were freed from etching from the top to the center. Cone-type structures are formed by etching of 1/3D of microspheres.

The optical characteristics of the PNJ generated by the obtained microstructures were investigated in the light transmission mode using a specially developed luminescent microscope, the detailed scheme of which is given in [10]. The wavelength of the exciting radiation was chosen as 532 nm due to possible further studies of the sensory response of rhodamine 6G based chemosensitive receptors [11], which luminescence excitation maximum is near the specified wavelength.

3. Results and discussion

3.1. “Crescent” microstructures

Figures 1a and 1b show the results of numerical modeling of the intensity distribution of the radiation passing through the crescent-type microstructures. A change in the microparticle surface profile (as compared to spherical [4]) leads not only to an increase in the length of the main PNJ, but also to the formation of the secondary one with lower intensity.

![Figure 1](image-url)

**Figure 1.** The parameters of the PNJ generated by “crescent” microstructures: a) the intensity distribution of a plane monochromatic wave passed through two microstructures located at a distance D; b) luminescent images of the main and secondary PNJ generated at 0 μm (1), 0.5 μm (2), 0.75 μm (3) and 1.5 μm (4) from the microstructure surface; c) and d) the intensity distribution of the main and secondary PNJs, respectively, along the propagation axis at different values of the environment refractive index.
An increase in the refractive index of the environment leads to a change in the conditions for the formation of both main and secondary PNJs (Figures 1c and 1d). In the case of water (n_c = 1.33), the length of the main PNJ increases, but its intensity remains comparable with the one in the air. With a further increase in the environment refractive index, the main PNJ generation region is shifted from the microstructure surface towards the propagation axis of radiation. It was found, for chitosan in the swollen state (n_c = 1.4) the distance from the surface of the microstructure to the localization region is about 150 nm, while in the case of polymer under normal conditions (n_c = 1.5) this shift is 1 μm (2λ). In addition, a gradual decrease in the main PNJ intensity, which, however, exceeds the noise level, was observed.

3.2. “Cone” microstructures
The cone-type microstructures also generate the secondary PNJ, and its form differs significantly from that presented earlier. In the air the secondary PNJ has two localization maxima (Figure 2a). The first, more intense, but having a smaller length (2λ), is formed immediately after the main PNJ. The length of the second maximum reaches 6λ, and it is separated from the first at a distance of 2λ. The intensity of the secondary PNJ in this case is significantly lower than for crescent-type microstructures and can hardly be distinguished by fluorescent microscope (Figure 2b).

![Figure 2. The parameters of the PNJ generated by “cone” microstructures: a) the intensity distribution of a plane monochromatic wave passed through two microstructures located at a distance D; b) luminescent images of the main and secondary PNJ generated at 0 μm (1), 0.5 μm (2), 0.75 μm (3) and 1.5 μm (4) from the microstructure surface; c) and d) the intensity distribution of the main and secondary PNJs, respectively, along the propagation axis at different values of the environment refractive index.](image)

In the case of water environment, there is a characteristic increase in the length of the main PNJ, accompanied by a decrease in its intensity (Figure 2c). One can note that with an increase in the
environment refractive index, the first intense maximum of the secondary PNJ disappears, while the second maximum remains (Figure 2d). It is characterized by lower intensity, but retains its length. A further increase in the refractive index of the environment leads to an increase in the main PNJ length and a decrease in the intensity of the secondary one. Compared to “crescent”, cone-type microstructures dispersed in the swollen chitosan demonstrate a similar length of the main PNJ, but at the same time, it has a smaller half-width and a greater intensity. It should be noted that for structures of the cone-type, no significant distance between the main PNJ and the microstructure surface was found, even for the case of chitosan under normal conditions.

4. Conclusion
Thus, the process of PNJ generation by alumina microstructures of different surface morphology dispersed in various media (n2n from 1 to 1.545) was investigated numerically and experimentally. It was found that the change in the surface profile has no less effect on the generated PNJ parameters than the ratio of the refractive indexes of the microstructure and the environment.

The crescent-type microstructures does not provide a sufficient intensity of the localization area. In turn, the “cone” microstructures reveal the effect of simultaneous generation of two PNJ types with high intensity and length (6λ). It has been proved by numerical simulation that such structures can be used to generate the PNJ in the optical chemosensor, providing high excitation efficiency of the sensor response. Using fluorescent microscopy, it is shown that PNJs have a high intensity and can be relatively easy registered. The PNJ generated by the cone-type microstructures have a high practical potential, since they satisfy two conditions: a high-intensity localization region with small sizes (main PNJ) and a localization region with the greatest possible length (secondary PNJ). It allows implementing two types of sensory systems, as are (I) for express analysis and (II) for highly sensitive measurements.

Acknowledgements

This work was financially supported by the Russian Science Foundation (project 18-72-00095).

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