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Peculiarities of the formation of an ensemble of photonic nanojets by a micro-assembly of conical particles

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Abstract. **We report the results of theoretical studies on spatially localised near-field light structures (photonic nanojets) arising during the scattering of laser radiation on a metasurface in the form of a single-layer ordered assembly of dielectric microcones embedded in a transparent matrix (silicone film). By solving Maxwell's equations using the method of computational electrodynamics, basic parameters of localised light structures (length, width, and peak intensity) are analysed in detail under the conditions of mutual influence of the light fields of adjacent microparticles. It is found that the main factors affecting the characteristics of photon nanojets in question are the spatial orientation of microcones and the degree of their embedding into the silicone matrix. It is shown that a number of spatial configurations of conical micro-assemblies allow the formation of an ensemble of photon nanojets with specific characteristics unattainable for isolated microcones.**

Keywords: *photonic nanojet, micro-assembly of particles, conical microparticle, near-field focusing.*

1. Introduction

The effect of optical radiation focusing near small material objects – the so-called effect of photonic nanojets (PNJs) [1, 2] – has been for long time and very successfully studied. The increased interest in this effect is stipulated by the prospects of its wide practical application in various fields of science and modern technology, for example, in three-dimensional (3D) nanolithography of surfaces [3], laser microsurgery [4], high-precision diagnostics of the parameters of dispersed media [5], etc. The very fact of the formation of a spatially localised high-intensity optical field region in the form of a light jet is a consequence of the aberration focusing of radiation by weakly absorbing micro-objects whose size is close to the wavelength of incident radiation (mesoscale particles). In contrast to the case of radiation focusing with an ordinary optical lens, the PNJ is formed in the near-scattering zone. We emphasise that it is precisely the mesoscale particle size that predetermines the size of the spatial region of PNJ existence, limiting it to the near zone of the light wave diffraction. Here, the optical field is characterised by the most complex spatial structure formed by a superposition of outgoing and evanescent modes, which allows for its more significant localisation than in the far diffraction zone.

Photonic (nano) jets may have a geometric shape both in the form of quasi-homogeneous narrow light filaments and spatially inhomogeneous regions of increased intensity with pronounced side lobes and secondary clots along the direction of radiation incidence [6, 7]. The PNJ specific spatial structure depends on the shape and microphysical parameters of the object that scatters radiation, and also on the type of radiation that illuminates the object (plane wave, focused beam).

It should be noted that in most studies relating to the control over the PNJ parameters, single isolated particles located in free space are commonly considered. The optical field distribution near such a particle only depends on the characteristics of incident radiation and the particle parameters itself. In this case, the use of a group of closely spaced micro-objects (micro-assembly) can significantly affect the parameters of individual nanojets, which is caused by the interference of optical fields in the diffraction of radiation on adjacent microparticles [8 – 10].

In practice, in order to lock the microparticles within the specified spatial zones, an organised array of microparticles is usually placed either on the surface of the irradiated sample [11 – 14] or on an auxiliary transparent substrate, for example, on a quartz plate [15] or a polymer film [16, 17]. Obviously, the use of not a single microparticle, but a group of such particles embedded into the locking matrix, may change the PNJ parameters from individual particles due to emerging collective effects, and also as a result of the scattered field interaction with a substrate.

The effect of an absorbing substrate on the parameters of an optical field resulted from the ensemble of seven microspheres was earlier theoretically studied in work [18], where it was established that the contribution of both the substrate and adjacent particles may by several times change the optical field intensity within the focal zones compared to the case of an isolated sphere in vacuum. The PNJ elongation and the shift of its maximum towards the surface of the parental spheres arranged as a hexagonal cluster were revealed by Pikulin et al. [19]. In our work [20], we theoretically investigated the main parameters (length, width, focal distance, and intensity) of photonic nanojets formed on the ordered single-layer arrays of transparent glass microspheres. It was found that the degree of manifestation of collective effects in the formation of an array of nanojets depends on the spatial structure of the ensemble of microspheres, their size, and repetition period.

The solution of a number of practical problems requires obtaining the most elongated PNJ. Traditionally, axicons are

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used to ensure such a modification of the light beam's focal waist, i.e. lenses in which one of the surfaces is conically shaped [21]. Characteristic feature of axicon focusing is a significant violation of homocentricity of the light beam incident onto the lens. Spatial structure of the optical field formed by such lenses contains an elongated and virtually diffraction-free focus region, the length of which depends on the apex angle (thickness) of the axicon.

It is currently reported on the technologies of producing ordered ensembles of microprisms [22], microcones [23], and also cubic objects (cuboids) [24] for the implementation of extreme focusing of optical radiation. However, the contribution of collective effects to the spatial and amplitude characteristics of the emerging PNJ in varying the distance between such particles, their size and spatial configuration of their placement in the substrate is not fully studied. A detailed study of basic PNJ parameters, namely, the length, width, and peak intensity, will reveal principal regularities of the PNJ ensemble formation, which opens up new prospects for their controlling.

This paper presents the results of numerical simulation of the near-field focusing of a light wave by an ensemble of microcones positioned at the nodes of a rectangular spatial lattice of a transparent silicone matrix. It is shown that, by varying the placement period and configuration of the assembly of microcones, it is possible to modify the parameters of emerging light nanojets in terms of increasing their intensity and length.

2. Numerical simulation technique

We consider the problem of a spatial structure of the near-field scattering of a monochromatic light wave on an ensemble of dielectric conical microparticles in a transparent film. This problem has a number of specific features. Firstly, the microcones are located fairly close to each other – at distances of the order of several radiation wavelengths, which entails the need to make provision for the interference of optical fields scattered by each particle of the micro-assembly. Secondly, the microcones are immersed into a matrix, the optical properties of which are different from those of the particles embedded into that matrix. Consequently, optical contrasts of the immersed and nonimmersed parts of a particle should be different, which gives a nontrivial pattern of wave diffraction on such a multicontrast structure.

To date, according to our data, there exist no analytical solutions or quantitative estimates for such light-scattering problems, except for the cases of optical wave scattering on a cluster of microspheres in a homogeneous environment [25] and also scattering on the nanoparticle aggregates in the approximation of effective permittivity of the medium [26]. In this paper we consider the problem of light radiation diffraction in a structurally inhomogeneous medium with scattering inclusions of mesowave dimensions, when allowance for the near-field collective effects becomes important. This circumstance predetermined the choice of the method of numerical integration of Maxwell's system of differential equations [27] for solving the problem posed.

We have considered the most common micro-assembly type representing a rectangular single-layer configuration of particles in a plane (Figs 1a, 1b). All particles in the micro-assembly are identical and have a conical shape with a height h and a radius a of the circular base. The array of microcones was placed inside a transparent substrate with a thickness $s \geqslant h$ and an arrangement period (lattice period) g, which corresponds to the typical case of self-assembly and the formation of dense ($g = a$) or sparse ($g > a$) packing of particles. The immersion depth f of microspheres into the matrix (Fig. 1c) could be varied in the range $0 \leqslant f \leqslant h$. It was assumed that the microcones were made of a glass nonabsorbing in the optical range and having a refractive index $n = 1.6$, while the substrate also represented a film of a nonabsorbing organic polymer having (for certainty) a refractive index $n_{\text{lay}} = 1.4$; the whole system was placed in the air ($n_0 = 1$).

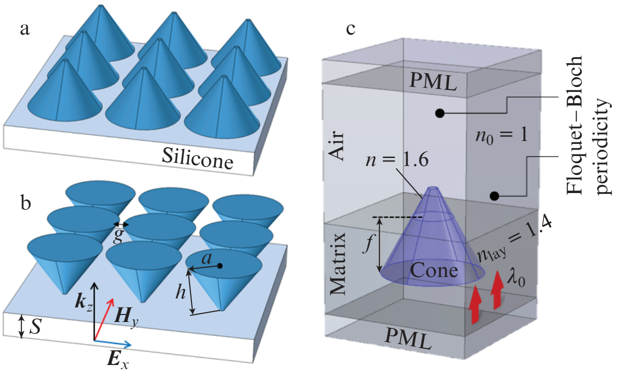

Figure 1. Schematic representation of the micro-assembly of cones in (a) direct and (b) inverse orientations; (c) 3D geometry of the computational domain used in numerical simulation.

Numerical calculation of the near-field structure in light-wave diffraction on the assembly of microparticles was conducted using the Lumerical FDTD Solutions commercial software package. A 3D geometry of the computational domain was used; herewith, due to spatial symmetry of the particle micro-assembly, the calculation was only performed for a single cell containing the particle and adjacent part of the matrix layer (Fig. 1c). The conditions of Floquet--Bloch spatial symmetry were set on the lateral boundaries of that region, while a set of perfectly matched layers (PMLs) was set along the direction of radiation incidence (z axis). The accuracy of numerical solution of the equations was controlled by an adaptive computational grid, the nodes of which were condensed within the areas of sharp gradients of permittivity of the medium (particle boundaries). The total number of grid nodes constituted several tens of millions, with spatial and temporal steps of the scheme ~2 nm and ~0.06 fs, respectively.

At the initial time moment, a source of monochromatic radiation representing a planar linearly polarised wave with a wavelength of $\lambda = 0.532$ μm and an electric field vector directed along the x axis was turned on at the lower boundary of the region. The optical wave propagated through the particle assembly in the direction of positive values of the z axis and formed a diffraction pattern in the near field of scattering, which was then averaged over a time interval of two picoseconds estimated to be sufficient to account for all the transients occurring in the particles and matrix. The thus obtained spatial distributions of the relative field intensity $B(x,z) = |\boldsymbol{E}(x,z)|^2/E_0^2$ (E_0 is the incident wave amplitude) were analysed to detect localised areas of increased intensity (PNJ regions) and determine their size and amplitude characteristics. The cases of direct and inverse orientation of particles in varying the depth of particle immersion into the substrate and the distance between individual microcones were considered.

3. Discussion of simulation results

First of all, we should note that the degree of packing density of particles is determined by the ratio of the parameters a and g, i.e. the closer the cones are positioned to each other, the more significant are the collective effects in the PNJ formation. We were interested in how the spatial configuration of the cluster of cones affects the parameters of generated photon jets.

We start from consideration of the effect of spatial orientation of microparticles in the locking substrate. Spatial distribution of the relative intensity B of the light field was calculated for the two cases of direct and inverse particle orientation with respect to the direction of its illumination. If a microparticle is oriented by a wide face towards the incident radiation (Fig. 1a), then this configuration is called the direct orientation. The case of an inverted cone (Fig. 1b) is called the inverse orientation. Figure 2 shows examples of the PNJ formation near single (adjacent particles are located at a distance $g \gg a$) glass cones with a base radius $a = 2\lambda$, when radiation with $\lambda = 0.532$ μm is incident onto them. The depth of cone immersion into the matrix is $f = h/2$.

In accordance with work [28], for the study and comparison of the PNJ from various particles, we choose the effective length L and transverse size R as basic parameters. These characteristics are determined in Fig. 2b when considering the spatial intensity profiles of the optical field in external focus region of a glass micro-axicon. We define the width and spatial extent of PNJ at the level of half the field intensity maximum (FWHM). The PNJ amplitude characteristic represents the magnitude of relative peak intensity $B_{\max}$ of optical field in the near scattering zone. The direction of radiation incidence is indicated by an arrow. The upper boundary of the silicone matrix is indicated by a solid line.

It can be seen that in both cases, spatially localised optical structures are formed near the shadow surface of particles, i.e., within the photonic nanojet region. However, for the case of direct cone orientation (Fig. 2a), the optical field intensity maximum is located inside the particle, while the PNJ itself is formed by a field 'flowing' from the focal waist. This stipulates an extremely small jet length ($L \approx 0.1\lambda$) and a not high intensity ($B_{\max} \approx 11$). Changing the spatial orientation of a particle from direct to inverse (Fig. 2b) leads to a sharp increase in the photon flux length outside the particle bounds. Now the intensity maximum is located out of the particle surface in the PNJ zone characterised by a larger amplitude ($B_{\max} \approx 35$) and a greater length ($L \approx 2\lambda$). This circumstance represents a direct consequence of a change in the optical contrast of the medium when a light wave crosses the boundaries of differently oriented particles [29].

Indeed, consider this situation from the standpoint of geometric optics under the assumption of large particles ($a \gg \lambda$). If the axicon is in direct orientation, its lateral surface, due to strong internal reflection close to total reflection ($n > n_0$, n_i), acts like a mirror for the rays coming inside through the planar base. As a result, light rays are collected mainly at the axicon top and come out at large angles to the optical axis, which limits the external focus region and prevents further concentration of light energy.

In the case of opposite orientation of the particle, rays fall on the cone lateral surface from the medium with a lower refractive index, so that their refraction occurs with a small reflection. After exiting through the cone base, the rays from each annular zone are collected on the axis at different distances and thus form an elongated focusing region typical of the axicon.

Understandably, within the framework of wave optics, the focusing model discussed above looks somewhat different, since diffraction effects come into play, which, in particular, leads to a more expressed optical field 'outflow' through the 'direct' axicon's top due to the appearance of decaying (evanescent) waves. However, in a whole, the described regularities also persist at mesoscales.

Next, by the example of inversely oriented axicon, we show how the PNJ parameters change at various degrees of the particle immersion into the polymer substrate. Figure 3 presents a spatial intensity distribution for the cases when the

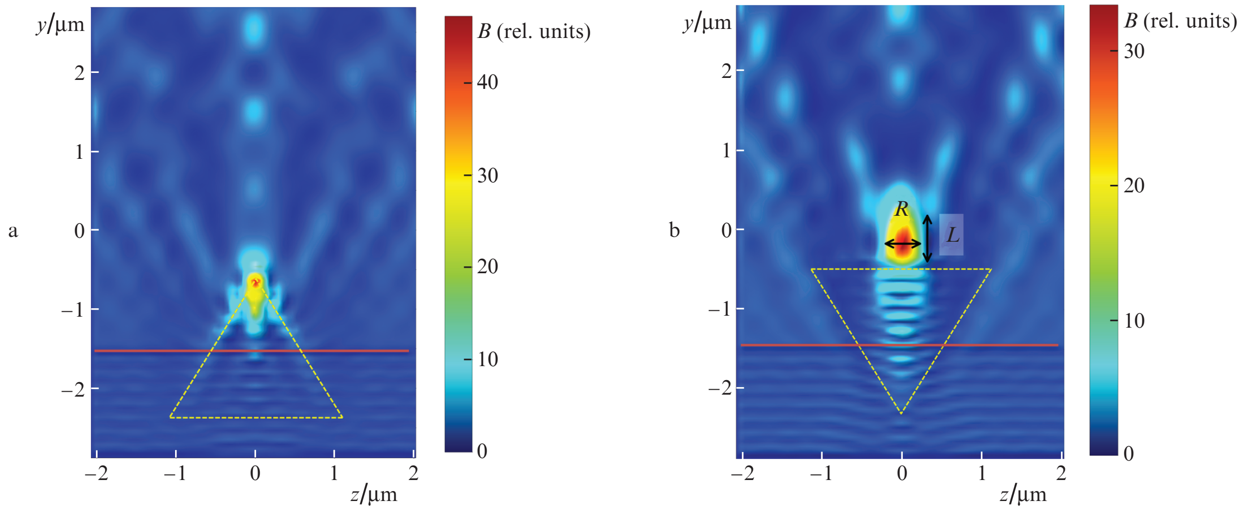

Figure 2. Spatial profile of the relative intensity B of the optical field in the vicinity of glass cones with a base radius $a = 2\lambda$, immersed into a polymer matrix for (a) direct and (b) inverse orientations (radiation is directed from the bottom upwards).

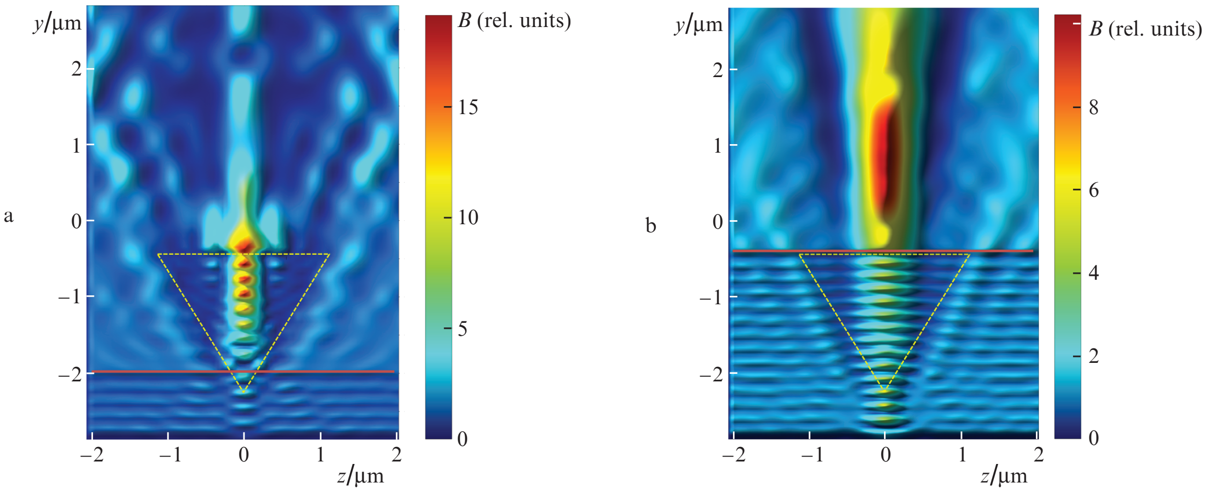

Figure 3. Spatial profile of the relative intensity B of the optical field in the vicinity of glass cones with different degrees f of immersion into the polymer matrix: f/h = (a) 0.125 and (b) 1. Radiation is incident from below.

particle is embedded into the matrix at various depths f. Here, as in Fig. 2, the upper boundary of the polymer film is marked by a horizontal line.

It follows from these Figures that, in contrast to the case of a partially immersed axicon (Fig. 3a), the complete immersion of a microparticle into the matrix (Fig. 3b) radically changes the spatial shape of the photon jet being formed. First of all, the PNJ length L is multiply increased, thus reaching $\sim 8\lambda$. Obviously, this is due to the focal waist elongation with a decrease in the numerical aperture of the particle-lens immersed into the medium with a refractive index greater than that of the air ($n = 1.4$), just as it occurs in the gradient-inhomogeneous Luneberg lenses [30, 31]. Numerical calculations also show that a sharp increase in the PNJ length is accompanied by an increase in the jet transverse size ($R \approx 2\lambda$) and also a drop in the peak intensity ($B_{\max} \approx 10$). For comparison, the corresponding values of the jet parameters for an axicon minimally immersed into the holding matrix (Fig. 3a) are as follows: $L \approx \lambda$, $R \approx 1.2\lambda$, $B_{\max} \approx 18$.

The simulation results presented below (Figs 4–6) display basic characteristics of the FNS ensemble in varying the distance g between individual particles and the degree f of the cone immersion into the matrix. For convenience of analysis, the dimensional parameters of the cone clusters and the PNJ produced by them are given in units of the radiation wavelength illuminating the particle. This allows us to generalise the established regularities, since, according to the Mie theory [32], the distribution structure itself of the optical wave field diffracted on the particle depends on the dimensionless R/λ ratio. We have considered the situations when the distance ratio g/a between individual particles in the cluster was varied from 0 to 1, which corresponds to different types of cone packing – from dense to sparse, respectively. As above, the degree of particle immersion into the substrate was regulated by the parameter f ($f/h = 0$ stands for a particle on the surface, $f/h = 1$ means the total immersion into the matrix).

As follows from Fig. 4, in the case of inverse particle configuration in the matrix (Fig. 4b), the dependences $L(f)$ in varying the distance g between individual particles are similar. It can be seen that at $f/h < 0.75$, the photon flux length does not depend on the degree of microcone immersion into the matrix. In other words, an ensemble of microcones partially immersed into a transparent silicone matrix forms a PNJ set of a certain fixed length regardless of the relative position of particles in the cluster. The nearest environment of each cone inversely oriented relative to the incident radiation affects the length of the PNJ formed in the near-field scattering zone only in the case of particles completely immersed into the substrate. Thus, the L value for densely packed particles ($g/a = 0$) is almost two times less than the corresponding value for sparse ($g/a = 1$) packing of cones,

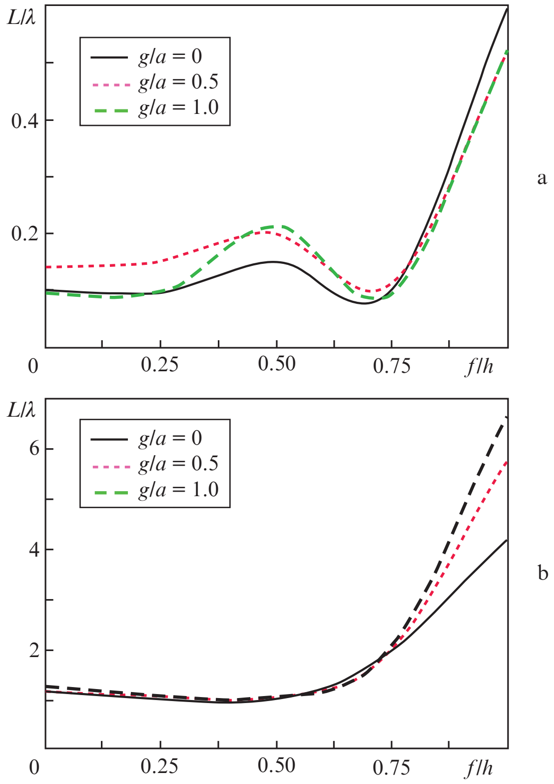

Figure 4. PNJ length for (a) direct and (b) inverse orientations of a cluster of cones in varying the depth f/h of immersion into the matrix and the distance g/a between the particles.

which is explained by the contribution of collective effects to the PNJ ensemble formation by the micro-assemble of conical particles.

A similar tendency is observed for direct orientation of conical particles on a substrate (Fig. 4a). Here, just as for inversely oriented cones, the main factor affecting the parameter L is the degree of particle immersion into the nonabsorbing matrix. The peculiarity of such focusing of the optical field by particles (see Fig. 2a) stipulates an extremely small jet length, since the optical field's maximum intensity lies predominantly inside the scattering particle. It is also important to note that the effect of adjacent microcones on the PNJ length is insignificant.

An important PNJ parameter is the transverse size R, which in the scientific literature is often called the spatial resolution of a photonic jet, which reflects the practice of using this parameter in ultra-high resolution microscopy [33] and precision detection of various nano-objects [34]. We show how the width of the light nanojets being formed is modified by varying the configuration of the assembly of micro-cones.

Let us consider Fig. 5. First and foremost, we should note that the similarity of dependences in the case of direct spatial orientation of microcones (Fig. 5a) indicates the invariance of the PNJ width in relation to the parameters of the assembly of conical particles. It can be seen from Fig. 2a that optical field localisation in directly oriented particles occurs in a limited region located at the cone apex. The resulting PNJ actually represent an outflowing optical field, which stipulates their smallness in length and subdiffraction transverse size. It is of interest that no noticeable change in the light nanojet width ($R \approx 0.38\lambda$) is observed when the microcones with direct orientation are immersed into the substrate. At the same time, for inversely oriented particles (Fig. 5b) the photonic flux width increases with the degree of immersion of microcones into the matrix. It can be noted that in a whole, the narrowest PNJs arise for partially immersed particles ($f/h < 0.50.5$).

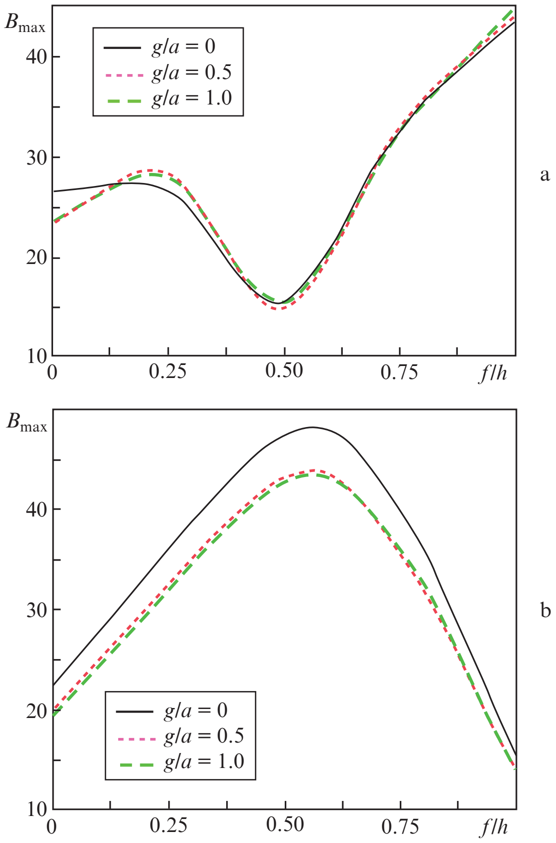

Figure 5. PNJ width for (a) direct and (b) inverse orientations of a cluster of cones in varying the depth f/h of immersion into the matrix and the distance g/a between the particles.

Figure 6 shows the results of numerical calculations of the peak values for the third important PNJ parameter, namely the maximum increase in relative intensity of the optical field B#max for the particle cluster cases under consideration.

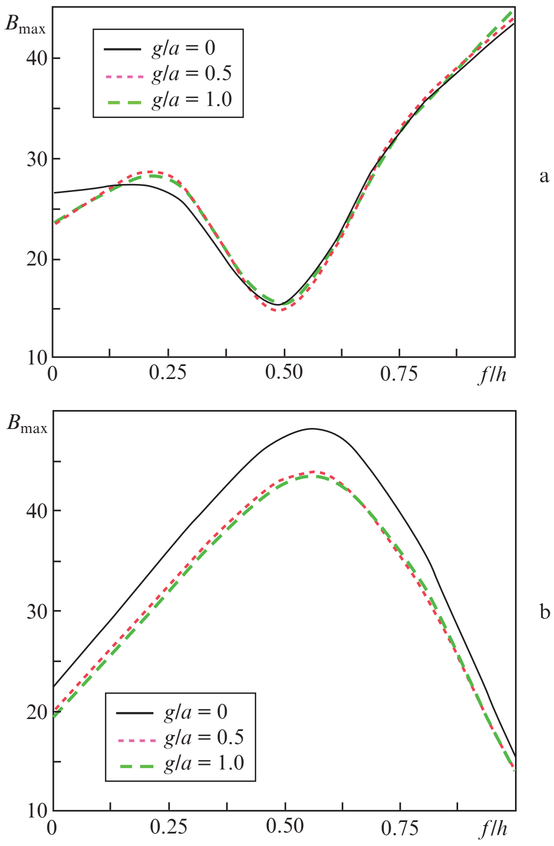

Figure 6. Peak intensity B_{max} of the optical field in the PNJ region for (a) direct and (b) inverse configurations of a cluster of cones in varying the depth f/h of immersion into the matrix and the distance g/a between the particles.

It can be seen that this parameter is characterised by a similar type of dependences for all variants of particle packing (dense or sparse). The jet intensity increases markedly (more than twice) in the case of inverse orientation with partial immersion of microcones into the substrate ($f/h \sim 0.5$). In the case of direct orientation of microparticles the effect is opposite: the peak intensity B_{max} drops sharply. This is caused by different deformation of the radiation wave front incident onto the particles, which entails a change in the focusing properties of the microcones.

4. Conclusions

Using the theoretical modelling of a spatial structure of the near-field of light wave scattering on the ordered cluster of dielectric microcones embedded into a transparent matrix, we have studied the characteristics of regions with high optical field concentration – photonic nanojets. The length, width, and intensity of the formed PNJ have been investigated for various orientations and spatial configurations of the particle placement in the substrate.

It is shown that each of the studied types of clusters is capable of forming a PNJ array with subwavelength spatial resolution in the near diffraction zone. The contribution of adjacent particles in this case is associated with the mutual interference of optical fields, and is manifested mainly in the pulsations of intensity and length of a photonic jet when the cluster lattice period changes. In this case, the most strongly varying are the PNJ parameters from an ensemble of particles whose cone apex is oriented against the direction of radiation incidence (inverse orientation). Direct orientation of cones is characterised by the greatest stability of the PNJ spatial parameters (length and width), whereas the power characteristics of the jets formed are very sensitive to the spatial placement of particles in the substrate, determined by the particle embedding parameter.

It has been established that, due to the interference of the fields of adjacent jets, a denser packing of inversely oriented cones inevitably leads to the pulsations of their intensity and may cause a spatial ‘rupture’ of the initially continuous PNJ into separate segments along the vector of optical wave propagation, which is expressed in the jet width fluctuations. As the distance between the particles increases, the contribution of collective interference effects decreases. It has also been found that the immersion of an ensemble of inversely oriented microcones into the polymer substrate to a depth equal to half the diameter of their base allows a more than twofold PNJ intensity increase regardless of the type of particle packing (dense or sparse).

For microparticles with direct orientation, of interest is the case of complete immersion of microcones into a matrix with a different refractive index. The thus formed PNJs have a very limited size, but high intensity and stably small transverse size of the jet.

In addition, with a certain arrangement of particles in the cluster, it becomes possible to implement the PNJ with parameters being significantly better compared to the case of an isolated cone. This represents a fundamental point in the implementation of a number of practical tasks. In particular, the diffraction interaction of fields from individual microcones can extend the tail part of the jet, as in the case of full immersion into the polymer matrix of an array of densely packed particles (direct orientation), or significantly reduce the transverse size of the jet with a corresponding increase in its intensity, as in the case of microcones partially immersed into the substrate (inverse orientation).

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