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# Characteristics of photonic nanojets from ordered microassemblies of dielectric spheres

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***Abstract.*** **Spatially localised light structures (photonic nanojets) formed in the near field of light scattering from an ordered ensemble of single-layer clusters of transparent glass microspheres have been theoretically studied. The main parameters of photonic nanojets (length, width, focal distance and intensity) are calculated based on numerical solution of Maxwell equations, and their behaviour under conditions of interference between neighbouring microparticles is analysed. The degree of manifestation of collective effects during the formation of a nanojet array is found to depend on the spatial structure of the ensemble of microspheres, their size and repetition period. It is found that in some cases, having chosen an appropriate configuration of a microsphere ensemble, one can improve significantly the characteristics of the photonic nanojets formed by this ensemble.**

***Keywords:*** *microsphere, light scattering, photonic nanojet, cluster of microparticles.*

## 1. Introduction

Recent advances in optical technologies have given rise to new promising lines of research, related to ultrahigh-resolution microscopy, nanooptics, nanodesign of materials, and precise diagnostics of parameters of dispersed media [1–4]. Most of these problems call for the formation of concentrated light beams near material objects of different physical nature. To date, several ways to obtain ultrahigh nanoscale localisation of an optical field have been developed. They include, in particular, excitation of surface plasmon resonances in nanoparticles [5] and application of nanoantennas [6] and optical tips [7]. However, all these techniques are based on the use of metal objects, which are known to strongly absorb light; this absorption causes significant heating (undesirable in some cases) of the samples.

In this context, researchers and technologists pay much attention to nonabsorbing dielectric microscopic objects with different geometric shapes, physical properties and structural compositions [8–14]. The use of these mesoscale microscopic objects [15, 16], i.e., objects with a characteristic spatial scale on the order of light wavelength, to localise light energy in very small volumes is fairly promising. Diffraction of electromagnetic radiation from microparticles leads to the formation of highly localised regions of enhanced intensity [the so-called photonic nanojets (PNJs)] near their surface [9, 17]. The PNJ nature is related to the near-field aberration focusing of light by a transparent particle [18]. In this case, the constructive interference of the fields of the scattered light and the light transmitted through the particle provides conditions for forming an ultrasmall (up to subdiffractive) elongated high-intensity focal region in the shadow zone.

It should be emphasised that the use of mesoscale particles provides strong spatial localisation of the electromagnetic field near small objects. In turn, this fact determines the spatial domain of PNJ existence, limiting it by the boundaries of the near-field zone of light wave diffraction, where the optical field is characterised by the most complex spatial structure and, in principle, can be localised more strongly than in the far-field diffraction zone.

In practice, manipulation of localised light fluxes formed by isolated microparticles is a rather complex problem, because there are some technical problems with fixation of suspended microparticles in space (if they are exposed to light) in order to affect selectively an object under study. In this context, the most widespread way to form and apply PNJs is to place an ordered array of microparticles (generally, an ensemble of transparent microspheres) either directly on the surface of an irradiated sample [19–23] or on an auxiliary transparent substrate (e.g., a polymer film [24, 25] or a quartz plate [26]). This approach allows one to fix reliably microparticles in desired zones and exclude their displacement from the initial position. Using this technique, one can implement high-precision micro- and nanoprocessing, punching of microholes and laser etching of various materials [1, 19, 20, 22, 25, 27], and detection of ultralow amounts of materials [23] and individual nanoparticles [24].

The most important problem of the PNJ optics is the control of jet characteristics. To date, several different ways to control the PNJ parameters and shape in order to increase the jet spatial resolution and intensity have been discussed in the literature [28–30]. The point is that the photonic jet parameters are fairly sensitive to variations in size, optical properties, structural composition, and geometric shape of the parent microparticle [31]. It was shown that subwave focusing of an optical field using a PNJ formed by microspheres (in particular, the generation of a light beam with a transverse width smaller than the diffraction width) can be implemented only in a rather narrow range of reduced sizes of spherical microparticles ($5 < x_a < 30$), i.e., for mesoscale particles. Here, $x_a = 2\pi D/\lambda$ is the reduced particle radius (Mie parameter), $D$ is the geometric size, and $\lambda$ is the incident light wavelength. For smaller particles, the spatial localisation of the optical field is still insufficient, whereas for larger particles the focal region has too large (atypical of PNJ) sizes.

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Note that most of the studies on the control of photonic jets considered generally single isolated particles in a free environment. The optical field distribution near an isolated particle depends only on the characteristics of incident light and the microphysical parameters of the particle. Obviously, the use of a group of microparticles (incorporated into a fixing matrix or placed on a substrate) rather than a single microparticle in order to form PNJs may affect significantly the parameters of individual nanojets due to the interference of optical fields during light diffraction from neighbouring particles and interaction between the scattered field and substrate.

This problem was discussed in [32–34]. The characteristics of the local light focusing regions formed by one- and multilayer clusters of identical microspheres were considered in [32]. It was shown that the strongest field focusing is observed near the particles forming cluster boundaries, because the optical field is partially redistributed from the central microspheres to the cluster periphery. The influence of the absorbing substrate on the parameters of the optical field formed by an ensemble of seven microspheres was theoretically analysed in [33], where it was found that both the substrate and neighbouring particles reduce several times the optical field intensity in the focal regions in comparison with an isolated sphere in vacuum. Elongation of a PNJ and shift of its maximum to the surface of parent spheres, arranged into a hexagonal cluster, was observed in [34].

A conventional way to form a PNJ matrix is to use a so-called self-assembly of particles during their arrangement on a surface studied or on a substrate. Generally, this process is related to the precipitation and evaporation of microparticles from a colloidal solution deposited on a substrate, with subsequent formation of a single- or multilayer matrix of closely packed particles [22]. Specifically these clusters of microspheres were considered in the aforementioned theoretical studies.

A specific feature of the close packing of particles is a quite definite mutual arrangement of the PNJs formed in it. For example, when microspheres are used, photonic fluxes are located at the vertices of equilateral triangles with a side equal to the diameter of an individual sphere, which form a periodic hexagonal structure on the surface [19]. This circumstance imposes an essential limitation on the control of PNJ configuration. Indeed, in the case of close packing, one can change the distance between individual PNJs by changing only the size of microspheres, which, in turn, inevitably leads to changes in the characteristics of photonic nanojets produced by these microspheres [8, 31].

At the same time, it was reported recently about the formation of transparent polymer (PDMA) films with incorporated glass microspheres and their application in ultra-high-resolution microscopy [35]. A unique feature of these ‘sphere-containing’ silicone films is that the microparticles incorporated into them can in principle be fixed in any spatial configuration rather than form only a close packing. Then a flexible polymer matrix can be transferred onto any surface to implement subwave and even subdiffraction light focusing [25]. In this context, an urgent problem is to study the collective diffraction effects occurring during PNJ formation from ensembles of differently arranged particles.

In this study, we considered the diffraction of an optical wave from a single-layer ensemble of identical microspheres, inserted in a matrix with a refractive index different from that of the spheres. Using a method of computational electrodynamics, based on numerical solution of differential Maxwell equations, we investigated (for the first time, to the best of our knowledge) the spatial and amplitude characteristics of localised photonic fluxes with variation in the distance between individual spheres, their size, and spatial arrangement in the matrix (square and hexagonal lattices). It turned out that the influence of the matrix (into which spheres are incorporated) manifests itself mainly in the elongation of a PNJ and decrease in its intensity. The collective interaction of the fields of a PNJ ensemble from an array of particles leads to highly unstable behaviour of PNJ parameters at distances between individual spheres smaller than several incident light wavelengths. Some spatial configurations of particles are characterised by the formation of PNJs with better parameters than in the case of isolated spheres.

## 2. Numerical simulation method

We considered the two most widespread types of microsphere assemblies: hexagonal and square single-layer configurations of particles in a plane (Fig. 1). The spherical particles were assumed to be identical, having diameter $D$ and arrangement (lattice) period $d$. An ensemble of particles was inserted in a dielectric matrix with a thickness $s \geqslant D$. The version with a hexagonal particle arrangement corresponds to the typical case of a self-assembly and formation of a close packing of microspheres. When particles are located at square vertices, the two-dimensional phase diffraction grating is, e.g., an ordered array of microsteps [26].

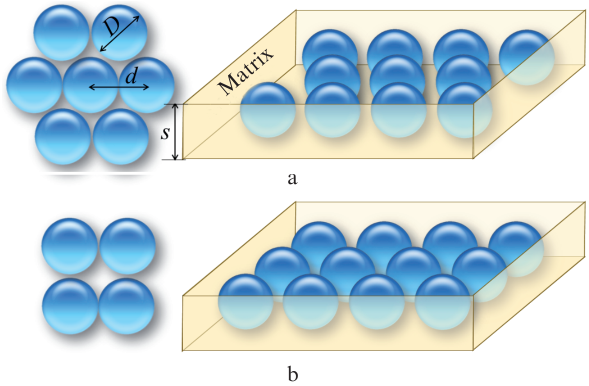


**Figure 1.** (a) Hexagonal and (b) square configurations of microsphere assemblies.

A numerical calculation of the near-field structure for the light wave diffraction from assemblies of microspheres was performed using a software package developed by us, having a core based on a $C^{++}$ module of three-dimensional FDTD with an open code (web resource [36]), modified for specific calculated configurations. We considered a three-dimensional configuration of the calculation domain, in which the profile of permittivity $\varepsilon$ along the coordinate axes was set in correspondence with the specific type of the ensemble. The microspheres were assumed to be made of transparent optical glass of heavy flint type ($\varepsilon = n^2 = 3.4$) with a low chromatic dispersion in the optical wavelength range. Assemblies of spheres were placed in a nonabsorbing silicone matrix with a permittivity $\varepsilon_f = 1.96$; the matrix, in turn, was placed in air ($\varepsilon_0 = 1$).

The geometric sizes of the calculation domain were varied, depending on the parameters of the problem, and could reach $15 \times 15 \times 20$ μm at a total number of computational grid points on the order of $10^8$.

To minimise the edge effects when simulating a hexagonal configuration of microspheres, we set a cluster composed of 25 particles (arranged in five rows) and investigated the PNJ parameters at its centre. The square arrangement of particles made it possible to simulate an infinite lattice of microspheres by applying periodic boundary conditions at the boundaries of the calculation domain along the $x$ and $y$ axes. A set of absolutely matched layers was specified along the $z$ axis.

A source of monochromatic light with a wavelength $\lambda$ in the form of a plane linear polarised wave with an electric field vector directed, for definiteness, along the $x$ axis was switched on at the lower boundary of the domain at the initial instant. An optical wave propagated through an assembly of particles in the positive direction of the $z$ axis and formed a diffraction pattern in the near scattering field, which was then averaged for 2 ps; this time was estimated to be sufficient to take into account all transition processes occurring in the spheres and matrix. The thus obtained spatial distributions of the light field relative intensity $B(x,z) = |\boldsymbol{E}(x,z)|^2/E_0^2$ ($E_0$ is the incident wave amplitude) were analysed in order to detect localised enhanced intensity regions (PNJs) and determine their size and amplitude characteristics.

## 3. PNJ from a single particle

Figure 2 shows examples of PNJ formation near single spherical particles with a diameter $D = 11\lambda$, exposed to light with $\lambda = 0.532$ μm. The spatial distribution of light field relative intensity was calculated for two cases: a particle inserted in a matrix (Fig. 2a) and a particle suspended in air (Fig. 2b).

It can be seen that intense spatially localised optical structures are formed near the external shadow surface of spheres in both aforementioned cases. Conventionally, we will refer to these structures as photonic nanojets (indicated in Fig. 2). The shape, intensity, and size parameters of PNJs are different and depend on the properties of the particle environment. For example, for a microparticle located in air, the optical field intensity maximum, as can be seen in Fig. 2b, lies in the sphere, and the PNJ zone is formed by the field 'flowing out' of the focal waist. These factors are responsible for the jet small extension (on the order of light wavelength) and low intensity.

The insertion of a sphere in a matrix with a refractive index higher than that of air, $n_f = \sqrt{\varepsilon_f} = 1.4$ (Fig. 2a), changes radically the PNJ shape. It is now located at a much larger distance from the particle surface and is characterised by a larger field amplitude and length (about $8\lambda$). This circumstance is a direct consequence of the decrease in the optical contrast of the medium for a wave intersecting the particle boundaries, which decreases the numerical aperture of the lens (spherical particle) and elongates the focal waist, as this occurs in gradient inhomogeneous spheres [29, 37].

Below, to characterise PNJs quantitatively, we introduce into consideration the size parameters of a jet as a localised light structure, specifically, length $L$, width $R$, and distance $f$ from the particle surface (the focal distance). The amplitude characteristic of PNJs is the peak (relative) intensity $B_m$ of the optical field within the near-field scattering zone.

To avoid ambiguity in interpreting parameters, we will determine the PNJ width at the level of half maximum of the field intensity and the length at a fixed intensity level: $B = 1$. The jet focal distance is calculated from the difference in the coordinates of the PNJ centre, where the light flux intensity is maximal, and the particle boundary. For complex estimation of the degree of spatial localisation of PNJ as a whole, we will use the quality criterion $Q$ [29], which includes three main jet parameters: $Q = B_m L/R$.

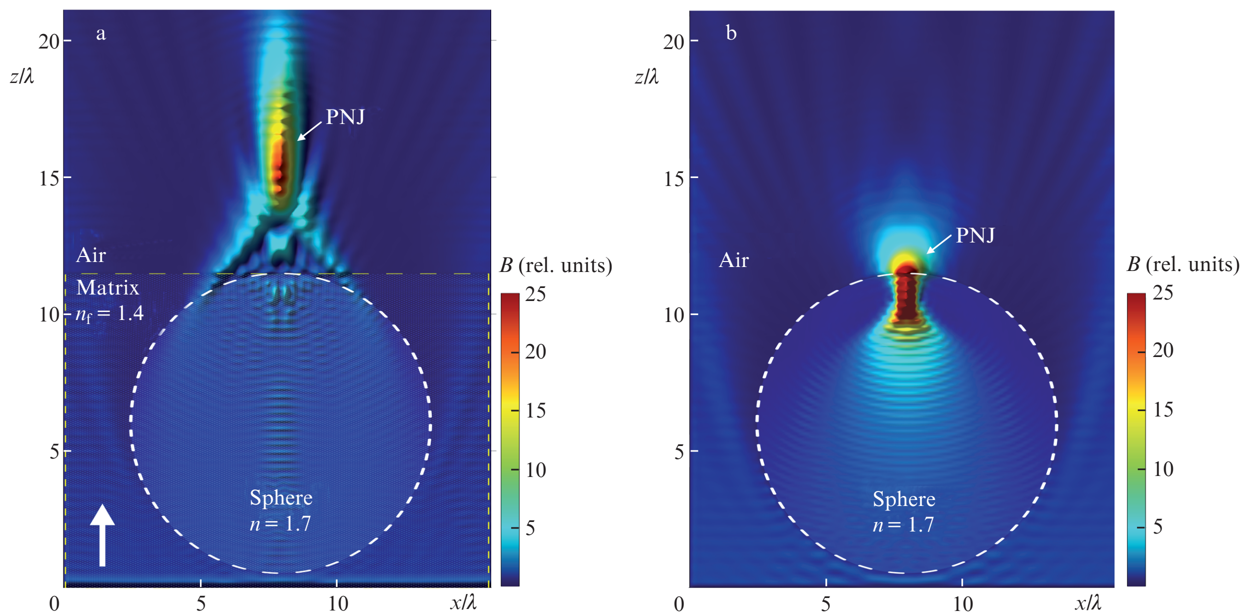


**Figure 2.** Spatial profiles of optical field intensity $B$ in the vicinity of glass spheres with a diameter $D = 11\lambda$, located in (a) a polymer matrix and (b) air. The arrow shows the incident light propagation direction.

## 4. PNJ from a cluster of closely packed microspheres

Let us begin consideration with PNJs formed by an ensemble of closely packed microspheres placed in a transparent matrix. As was mentioned above, the objects of this study are two spatial configurations of particle arrangement in a plane: hexagonal and square. The close packing of identical spheres implies validity of the relation $d = D$, i.e., close contact of neighbouring-particle surfaces in the horizontal and vertical directions. We intended to answer the following question: how do the configuration of a cluster of spheres and their sizes affect the parameters of generated photonic jets?

Figure 3 shows the results of the corresponding calculations. Here (and in other figures), all size parameters of the clusters of spheres and the PNJs produced by them are given in incident light wavelengths. This approach allows one to generalise the found regularities, because, according to the Mie theory [38], the structure of the optical field distribution for a wave diffracted from a sphere depends on the dimensionless ratio $D/\lambda$.

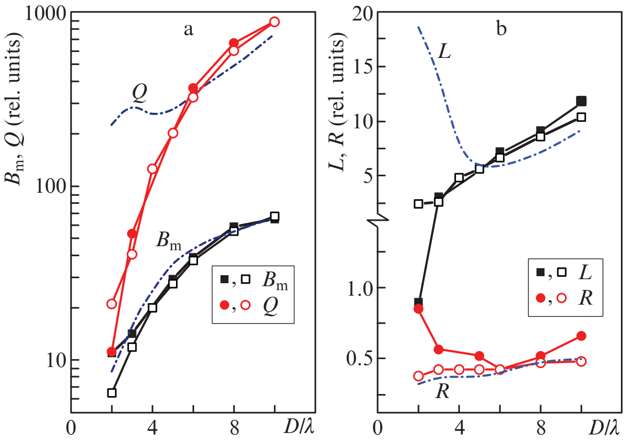


**Figure 3.** Parameters of photonic nanojets from clusters of closely packed microspheres of different diameters in (□, ○) hexagonal and (■, ●) square configurations. The characteristics of PNJs from single spheres are shown by dot-and-dash lines.

Thus, in the near-field scattering zone, as follows from Fig. 3, the configuration of closely packed particles affects only slightly the main PNJ characteristics. The only exception is the case of small spheres (with a radius of one wavelength), when the square lattice of particles is much worse (in terms of jet quality) than the hexagonal lattice. Primarily, the jet length sharply decreases and its width increases (Fig. 3b) for a square lattice, although the peak PNJ intensity is almost twice as high in this case (Fig. 3a).

In other cases, an increase in the sphere size leads to a monotonic increase in the photonic jet parameters, independent of the lattice configuration. The jet quality factors for two extreme values of particle diameter differ by almost two orders of magnitude. A subwave jet width is implemented in all cases. All PNJs are formed rather close (on average at a distance of $f = 1.5\lambda$) to the surface of spheres (not shown in Fig. 3).

It is of interest that the quality of PNJs produced by a cluster of particles and by a single sphere in the same polymer matrix differs for only small particles ($D \leqslant 5\lambda$). The main difference is the much larger length (up to several tens of wavelengths) of the PNJs from small single spheres. At the same time, if spheres form a closely packed cluster, the interference of the fields of neighbouring jets should inevitably lead to pulsations of their intensity and may cause separation of an initially continuous PNJ into several segments in the optical wave propagation direction. With an increase in the particle size, the influence of collective interference effects weakens both due to the increase in the distance between jets and as a result of a sharp increase in their intensity, which manifests itself in smoothing out longitudinal jet breaks.

## 5. Influence of the lattice period of a microsphere cluster

Below we present simulation results for an ensemble of PNJs, obtained with variation in the distance between individual microspheres in a cluster for the two lattice configurations considered here. It is more convenient to analyse these data using the intersphere gap $g = d - D$ rather than the particle lattice period $d$.

Figure 4 presents the main parameters of PNJs formed by an ensemble of spheres. As in Fig. 3, the reference values of the corresponding parameters of a jet formed by single particle are also given.

The most noteworthy thing is the high variability of jet characteristics with variation in the distance between parti-

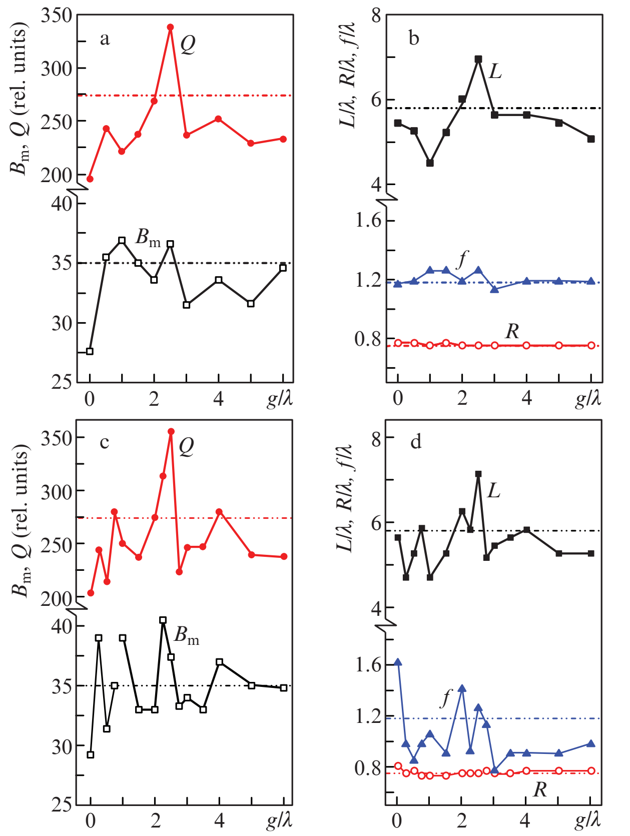


**Figure 4.** Dependences of the main PNJ parameters for the (a, b) hexagonal and (c, d) square configurations of a cluster of spheres ($D = 5\lambda$) on the intersphere gap. The dot-and-dash lines indicate the reference values of the parameters.

cles. This is most weakly pronounced for the jet width, whereas the corresponding values of intensity, length, remoteness, and total quality of PNJs may differ several times even at a small variation in the gap. The hexagonal configuration of spheres leads to a smoother dependence of PNJ characteristics on parameter $g$, in contrast to the square configuration. Obviously, this is a manifestation of the fact that the nearest environment of each sphere in the square lattice includes eight particles, whereas it has only six neighbours in the hexagonal lattice. The interference of the fields from a larger number of particles is also characterised by a higher sensitivity to a change in the intersphere gap. However, stabilisation of the jet parameters at the levels implemented for a single sphere occurs at approximately the same gap value for both lattices: $g \geqslant 4\lambda$.

An analysis of Fig. 4 shows that close packing of microspheres with $D = 5\lambda$ leads to the formation of a PNJ with the lowest quality factor. Some mutual arrangement of microspheres gives rise to a PNJ with the highest ($Q \approx 350$) quality factor. This situation is implemented at the same value of parameter $g$ ($2.5\lambda$) for both lattice configurations under consideration. Moreover, the parameters of this photonic jet are better than those of the PNJ from an isolated sphere of the same size, for which $Q \approx 275$.

Figure 5 shows a longitudinal profile of optical field relative intensity in the PNJ region for an isolated sphere and for a cluster of particles. The beginning of the jet corresponds to the relative coordinate $z/\lambda = 5.7$. It can be seen that the jet formed by a single sphere has an almost ideally smooth intensity profile with a maximum $B_m = 35.5$, spaced from the particle surface by approximately one wavelength, and an exponentially decaying tail. In this case, the PNJ length is little less than six wavelengths ($L \cong 5.7\lambda$).

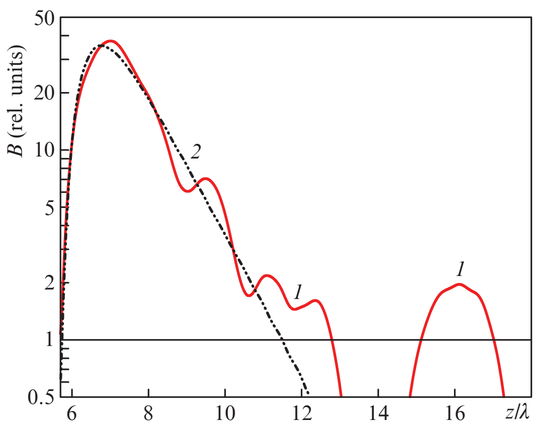


**Figure 5.** Optical field intensity profile in the vicinity of a PNJ (*1*) from a cluster of microspheres with $D = 5\lambda$ in a square configuration with $g = 2.5\lambda$ and (*2*) from an isolated sphere.

If a sphere is surrounded by similar particles, the diffraction interaction of the fields from individual microspheres can either suppress the PNJ (e.g., at $g = 3\lambda$) or, as in the case under consideration, elongate its tail. For a square matrix of spheres, the jet can be elongated, as follows from Fig. 5, at most by $\sim 1.5\lambda$, with a small increase in the peak intensity $B_m$ (to 37.4) in comparison with a single sphere. The PNJ does not stop after the first intensity minimum but demonstrates pulsating behaviour, characteristic of the interference pattern of the sum of coherent fields from spatially spaced sources.

Returning to Fig. 4, we should note that the field interference is also responsible for the following feature: even at rather large gaps between particles in a cluster ($g > 6\lambda$), the PNJs formed by them are, nevertheless, somewhat shorter than the photonic jet from an isolated sphere. Both the width and peak intensity of cluster-produced PNJs reach the reference levels for a single particle.

## 6. Conclusions

Based on the theoretical simulation of the near-field spatial structure for light wave scattering from an ordered cluster of dielectric microspheres inserted in a transparent matrix, we studied the characteristic of regions with high optical field density: photonic nanojets. The length, width, remoteness, and intensity of PNJs formed by particles in hexagonal and square configurations were investigated. All aforementioned PNJ characteristics were found to depend on the size parameters of spatial lattices of particles, which makes it possible to control photonic jets by varying the size, period, and configuration of microsphere clusters.

Each cluster type under study can form an array of photonic jets in the near-field diffraction zone with a subwave spatial resolution. The collective effects occurring during PNJ formation are related to the interference of optical fields of neighbouring particles and manifest themselves mainly in pulsations of photonic jet intensity and length with a change in the cluster lattice period. The parameters of PNJs formed by a square lattice of microspheres change most significantly, whereas the hexagonal cluster configuration is characterised by the highest stability of PNJ parameters to variation in the intersphere distance.

It was established that, on the whole, a cluster of microspheres forms photonic jets of worse quality in comparison with a jet from an isolated microsphere of the same radius, located in a matrix. At the same time, the influence of collective interference effects weakens with an increase in the particle size and interparticle distance, both due to the larger spacing between jets and because of the sharp increase in their intensity. Thus, the quality of PNJs from a particle cluster approaches the quality of a jet formed by a single sphere. On the other hand, a certain arrangement of microspheres in a cluster allows one to implement a photonic flux with a quality factor greatly exceeding that for an isolated microsphere. For spheres with $D = 5\lambda$, this situation corresponds to the arrangement of particles with a gap of one sphere radius, independent of the spatial lattice configuration.

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