



Generation of highly confined photonic nanojet using crescent-shape refractive index profile in microsphere

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ABSTRACT

Photonic nanojets (PNJs) owing to their sub-wavelength near-field features have found many interesting applications like nanoscopy, nano photolithography, high density optical storage, enhancement of Raman signal and single molecule spectroscopy etc. More recently, the focus of research has been on tailoring of PNJs either for better confinement and thus higher peak intensity or for elongation of nanojet for high resolution far field applications. In this paper, we show that crescent-shape refractive index profile (CSRIP) of microspheres can be used to generate highly confined PNJ. By optimizing the refractive index of different layers in CSRIP microsphere, we show a free space confinement down to $\sim \lambda/4.5$ (FWHM ~ 110 nm for excitation with 500 nm wavelength). Further, it was observed that the optical properties of substrates also modulate the PNJ characteristics and lead to a further improvement in the transverse confinement to $\sim \lambda/6.7$.

1. Introduction

Generation of photonic nanojet (PNJ) and controlled manipulation of its characteristics has attracted considerable current research interest. Photonic nanojet, characterized by high intensity, low divergence and sub wavelength lateral confinement, was first reported by Chen et al. in 2004 for the scattering of plane wave by lossless dielectric micro-cylinder and microsphere [1,2]. Generation of photonic nanojet is a non-resonant phenomenon and has been shown to be relatively insensitive to the deformation and surface corrugations in the microspheres [3,4]. Due to their unique characteristics, PNJ have found many applications in areas like Raman signal enhancement [5,6], single molecule spectroscopy [7], fluorescence correlation spectroscopy [8], nano photolithography [9–11] and nanoscopy [12,13] etc. Several approaches have been proposed for manipulating the length and confinement of photonic jet as many of aforesaid applications require control over the length and/or lateral dimension of the nanojet. The results reported in literature show that the length and full width at half maxima (FWHM) of photonic nanojet depend on size and shape of micro particle, refractive index of particle and that of the surrounding medium, wavelength, polarization and beam profile of the excitation light etc. [14,15]. Apart from conventional shapes like sphere and cylinder, particles of other shapes such as axicon [16], micro cuboids [17,18], micro discs [19], concentric graded index microsphere or ellipsoids [20,21], core-shell microspheres [22,23] and truncated microspheres [24,25] etc. have also

been explored in various studies to investigate tuning of PNJ characteristics. More recently Eti et al. have demonstrated controlled manipulation of photonic nanojet using controlled tuning of the refractive index in liquid crystals filled micro shells [26].

Wu et al. [9] have demonstrated mask-less lithography with a feature size of 250 nm ($\sim \lambda/1.6$) using a self-assembled planar structure of silica microsphere with a 400 nm centred ultraviolet (UV) broad band light source. McLeod and Arnold [10] have shown nano-lithography using photonic nano jet generated by optically trapped submicron size polystyrene microspheres with 355 nm laser source. A resolution of 102 nm ($\sim \lambda/3.5$) and 130 nm ($\lambda/2.7$), was shown for 0.5 μm and 0.8 μm sized polystyrene microspheres respectively. The approach followed by McLeod is however difficult to implement for practical applications due to inherent instability in trapping of small micro-spheres and associated Brownian motion. Using a bigger micro-sphere (\sim few micrometer diameter) can therefore be more useful for controlled nano photolithography if PNJ with narrow FWHM can be generated with such microspheres.

In this paper, we present the results of our studies on the variation of FWHM and length of PNJ generated from micro-spheres having crescent shape refractive index profile (CSRIP). The results show that PNJ with FWHM down to $\sim \lambda/4.5$ can be achieved in free space with multilayer CSRIP lossless dielectric microsphere of 3 μm diameter. In the presence of commonly used substrates with higher refractive index this would enables writing feature as small as $\sim \lambda/6$.

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2. Theory and simulations

A number of analytical and numerical approaches have been developed and applied to study the near as well as far field distribution of electromagnetic field in the photonic nanojet generated by microscopic dielectric objects. Analytical approach based on Mie theory and its suitable extension have been used to analyse the field distribution inside and outside the microsphere for concentric as well as eccentric inclusions [22,27–29]. However, with increasing number of inclusions the number of interfaces and boundary conditions make the derivation of analytical solution intractable. Among the numerical approaches, Finite Difference Time Domain Method (FDTD) [30] is a widely used technique that was first used to demonstrate the existence of photonic nanojet by Chen et al. [1]. However due to regular shape of grids (square/cubic element in 2D/3D) used in FDTD, it requires extensively large number of elements to capture the details of object with sharp or irregular feature such as at the apex of crescent in the present work. Discrete dipole approximation and multiple multipole method based approaches have also been explored to study the generation and characteristics of nanojet from dielectric objects [31–33]. In the present work, we have used finite element method (FEM) based software package COMSOL Multiphysics 4.2 for solving the Maxwell's equation in our simulations. The use of FEM enables handling the geometries with varying feature sizes that allows better representation of sharp crescent tips. The entire computational domain was meshed with triangular elements of varying sizes using the built-in non-uniform mesh option. It is important to note here that the maximum mesh size used must be sufficiently smaller than the typically recommended element size of $\leq \lambda/10$ to ensure numerical accuracy of the simulation results. The maximum elemental size of the triangle used in our simulation was restricted to 20 nm and finer meshes were used near the interfaces. Further refinement in the mesh size did not lead to any change in the results. All simulations were carried out with perfectly matched layer (PML) surrounding the region of interest to ensure that no back-reflection take place at the finite boundaries of computational domain. We used direct solver SPOOLES (Sparse Object Oriented Linear Equations Solver) which ensures the convergence of simulation by keeping fine meshing with relatively lesser memory requirement. Since the experimental measurements involve illumination with focused Gaussian beam the standard plane wave illumination used in COMSOL script was suitably modified. Following the convention used in Multi-physics module, we take the x-axis as direction of propagation and electric field oscillations were taken along z-axis. The variation of electric field as a function of propagation distance is given by the following definitions [23]:

$$E(x, y) = E_0 \sqrt{\frac{\omega_0}{\omega(x)}} e^{-(y/\omega(x))^2} \cos\left(-kx + \eta(x) - \frac{ky^2}{2R(x)}\right) \quad (1)$$

where $\omega(x)$, $\eta(x)$ and $R(x)$ are the beam radius as a function of x , the Gouy phase and the radius of curvature of the wave front respectively and are defined as:

$$\omega(x) = \omega_0 \sqrt{1 + \left(\frac{x}{x_0}\right)^2}; \quad \eta(x) = \frac{1}{2} a \tan\left(\frac{x}{x_0}\right)$$

and $R(x) = x \left(1 + \left(\frac{x_0}{x}\right)^2\right)$; with $x_0 = \frac{\pi \omega_0^2}{\lambda}$ being the Rayleigh range.

The beam waist was taken as $\omega_0 = 2 \mu\text{m}$ to ensure that the microsphere is fully illuminated. All simulations were carried out using direct solver for computing the field. To ensure convergence, the mesh size was refined till the variation in computed field at any given point is $< 10^{-5}$.

3. Results and discussion

Simulations were first performed for single microsphere of $3 \mu\text{m}$ diameter while varying the refractive index ' n ' of microsphere from 1.3 to 2.0. The excitation wavelength was taken as 500 nm and the

surrounding refractive index was taken as 1.0. A typical intensity distribution in the photonic nanojet and the variation of FWHM as a function of refractive index of micro-sphere are shown in Fig. 1. As the refractive index of microsphere increases the FWHM of photonic nanojet generated by microsphere decreases due to sharper bending of the photons at curved surfaces of microsphere resulting in better confinement. The smallest FWHM achieved was 204 nm corresponding to the refractive index 1.7. Further increase in the refractive index leads to the formation of PNJ that has peak intensity inside the microsphere. Thus the useable PNJ protruding outside the microsphere have larger FWHM. The transverse profile of nano jet (along the y-axis) for micro-sphere having refractive index 1.7 is shown in the inset plot.

In our recent study we have shown that a system consisting of core-shell microspheres allows generation of photonic nanojet with controllable length and confinement [23]. While the generation of highly elongated PNJs is possible when the refractive index of the core is less than that of the shell, better confinement is obtained when shell refractive index is lower than the refractive index of the core. These results can be best understood by closely observing the variation of pointing vector inside the microsphere [22]. It was shown that when an inclusion microsphere with lower refractive index is present, it counters the rapid divergence beyond the focus point thereby leading to an elongated region of high intensity in the nanojet. On the other hand, better lateral confinement of the nanojet is obtained when inclusion microsphere has higher refractive index than the host microsphere. As one would expect, in this case the inclusion microsphere with higher index leads to further focusing of the beam. The best lateral confinement is obtained with eccentric configuration when the inclusion microsphere touches the surface of the shell at the point of emission of nanojet forming a crescent shaped refractive index profile in the microsphere. The crescent structure under consideration consists of a microsphere with an eccentric inclusion. An inclusion sphere (radius ' r_1 ') with higher refractive index ' n_1 ' is embedded in a larger sphere (radius ' r_2 ') with lower refractive index ' n_2 ' as shown in Fig. 2(a). The best lateral confinement of PNJ is obtained when the centres of two microspheres are separated by $d = r_2 - r_1$. The variation of FWHM of PNJ as a function of position of inclusion microsphere is shown in Fig. 2(b). The radius and refractive index of the outer microsphere was taken as $1.5 \mu\text{m}$ and 1.3 and that of the inclusion microsphere was $1 \mu\text{m}$ and 1.7 respectively. To obtain the best possible confinement numerical simulations were performed by varying the refractive index of the two micro-spheres in the range 1.3 to 2.1 with a constraint that the refractive index of outer microsphere is kept lower than that of the inclusion as shown in Fig. 3. For a chosen refractive index of the outer microsphere, the refractive index of inner layer is varied in the above mentioned range of refractive index as long as the peak intensity region in nanojet is outside the microsphere surface. Note that the nanojet with peak intensity inside the microsphere will be of no interest for many applications. The optimum combination of refractive index obtained was 1.3 and 1.7 for the outer and inclusion microspheres respectively which leads to a PNJ with FWHM of 140 nm. The intensity distribution corresponding to this configuration is shown in Fig. 4(a).

Introducing a second microsphere with refractive index higher than the first inclusion lead to further narrowing of lateral dimension of the PNJ. For this, a microsphere with diameter $1 \mu\text{m}$ was placed inside the two layered CSRP structure discussed above and the refractive index of all three layers were varied from 1.3 to 2.1, keeping refractive index of innermost inclusion layer higher than that of the immediate overlaying crescent shaped layer and so forth. The smallest FWHM obtained for this geometry was $\sim 140 \text{ nm}$ ($\lambda/3.6$) for refractive index of different layers being 1.9, 1.6 and 1.3 starting from the inner most inclusion to the outer microsphere respectively. The Intensity distribution for this geometry is shown in Fig. 4(b).

We investigated the effect of further increase in the number of inclusions by introducing microspheres of diameter $0.5 \mu\text{m}$ and $0.25 \mu\text{m}$, resulting in up to five crescent shaped layers. The optimized refractive

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