Near field dielectric microlenses

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ABSTRACT

The electromagnetic backscattered response of a metallic nanoparticle located close to a dielectric microsphere illuminated by a plane wave or a focused beam is theoretically investigated. It is demonstrated that the main contribution of the microsphere consists in enhancing the excitation field which produces an enhanced plasmon response. Furthermore, investigation of dipolar emission close to the microsphere shows a redirection of the radiated field in the backward direction.

Keywords: Micro-optical devices; Mie theory; Electromagnetic optics; scattering, particles; *alexis.devilez@fresnel.fr

1. INTRODUCTION

Achieving strong concentration of light has become a challenging goal in the field of optics to increase and localize potential interactions between propagating electromagnetic field and localized energy states of matter. During the last century, light focusing has been widely performed with classical microscopes. But, because they operate in the far field regime, their resolution is diffraction-limited. In the past decades, metal nanostructures, such nanoholes [1,2] and nanoparticles [3-6] have emerged as efficient materials to overcome the limitations of far field optics and achieve strong confinement of the electromagnetic field using localized plasmon resonances. However, the improvement of the couplings between the far field domain and these structures remain of major importance.

Recently, dielectric microspheres have attracted a renewed attention for their ability to focus light in their near field when illuminated by an optical wave [7-13]. Although the size of the particle is on the order of the incident optical wavelength, the microsphere produces a beaming which can be employed to increase light/matter interactions. When the microsphere is illuminated by a plane wave, the so-called "photonic nanojet" beam that emerges from the sphere has subwavelength transverse dimensions and low divergence, which makes it fruitful for applications in dry laser cleaning [14,15], nanopatterning [16-18], Raman spectroscopy [19,20] and optical data storage [21]. In particular, these recent investigations have highlighted a strong enhancement of the backscattered power when a metallic nanoparticle is located within this so-called photonic nanojet beam [7, 8] enabling enhanced sensing of nanoparticles.

However, due to its large dimension along the optical axis (typically 2-3 μ m), the photonic nanojet does not provide three-dimensional subwavelength light confinement. For applications requiring high transverse and longitudinal resolutions, the classical photonic nanojet is unsuitable, as performs no better than the focusing obtained from a classical microscope objective with a high numerical aperture. Quite surprisingly, it has recently been shown that a single microsphere illuminated by a tightly focused beam can outperform classical microscope systems and significantly enhance the fluorescence emission from a single molecule [22-25]. In that case, strong confinement of light, on the order of $(\lambda/n)^3$, with a non-resonant dielectric structure was clearly demonstrated.

In this release, we investigate the optical modification produced by the location of a metal nanoparticle close to a dielectric microsphere. In particular, we study the ability of microsphere to enhance backscattering signal of a nanoparticles located in its vicinity. We underline the role of the dielectric microsphere to focus light but more interestingly to collect radiation scattered by the particle.

2. FOCUSING LIGHT WITH DIELECTRIC MICROSPHERES

The near field intensity pattern of beaming emerging from a single microsphere illuminated by a plane wave and a focused Gaussian-like beam have been displayed respectively in Figure 1a and Figure 1b. Simulations are performed in the framework of Lorentz-Mie theory [26, 27] which enables fast calculations of the electromagnetic field in spherical

coordinates (Mie expansion is truncated at $n_{max} = 30$). In another hand, this expansion in terms of multipole distribution can facilitate physical interpretation of the couplings between the incident beaming and the localized plasmon resonances [28]. The incident Gaussian beam employed in figure 2b as incident beam is simulated using Davis approximation of Gaussian beams with s = 0.3 [29]. Although this approach may be insufficient to describe the off-axis electromagnetic field, Davis approximation enables to investigate physical properties of focused beam especially on-axis.

In both figure 1a and 1b, the 2μ m diameter silica microparticle (n_s = 1.6) is surrounded by a water based solution (n₀ = 1.33) and illuminated at λ = 500 nm (wavelength in vacuum). This weak optical index contrast produces a both narrow and elongated beam for the plane wave incident field. Previous studies have highlighted that the intensity transversal cross section was fitting a Gaussian distribution while the longitudinal cross section was following a Lorentzian decay [13]. When the microsphere is illuminated by a highly focused beam [22-25], the longitudinal extend of the emerging beaming drastically drops compared to the beam displayed in Figure 1a. The relative position of the incident focal zone and the microsphere is of crucial importance. In Figure 2b, the incident beam is focused at the origin, 600 nm away from the microsphere surface. This shift enables high angular components of the incident field to pass aside the microsphere and interfere with the field that propagates through the sphere [23]. It results a three dimensional confinement of the field at the outside surface of the microsphere.

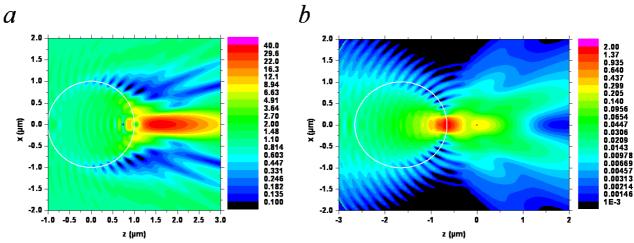


Figure 1: (a) Intensity colored map of a photonic jet beaming produced by a 2- μ m diameter sphere illuminated by a plane wave of wavelength in vacuum $\lambda_v = 500$ nm. (b) Intensity colored map of confined beaming produced by a 2- μ m diameter sphere illuminated by a tightly focused beam of wavelength in vacuum $\lambda_v = 500$ nm. The dielectric microsphere has a refractive index $n_s = 1.6$ and is surrounded by a medium of refractive index $n_0 = 1.33$.

3. PARTICULE IN PHOTONIC JET

A particle is now placed in the previously described beamings. Figure 2 displays intensity colored map of a 100 nm nanoparticle located (a) in the photonic jet beaming and (b) in the confined field produced by a Gaussian illumination on the microsphere. As the incident wavelength ($\lambda = 500$ nm) matches the dipolar resonance of the nanoparticle, one can notice that the maximum field in (a) and (b) is located on the surface of the nanoparticles and is several fold enhanced compared with Figure 1. Figure 2a shows that the photonic jet beam is still intense, revealing that the scattered field is mainly propagating in the forward direction. However, in both cases, the interference pattern inside the sphere betrays the enhancement of the backscattered power.

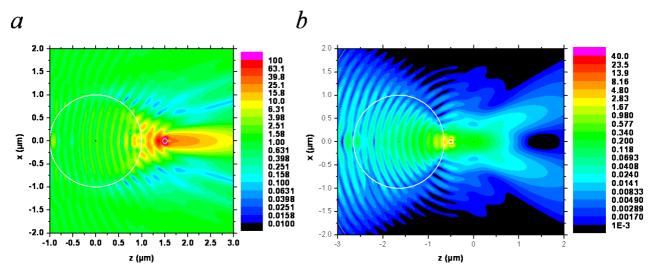


Figure 2: intensity colored map of a 100 nm nanoparticle located (a) in the photonic jet beamings and (b) in the confined field produced by the Gaussian illumination of the microsphere. The incident wavelength is $\lambda = 500$ nm.

As a first attempt to investigate the far field response of this system, the extinction cross-sections under plane wave illumination have been displayed in Figure 2a for the microsphere (2μ m in diameter and $n_s = 1.6$), Figure 2b for a 100 nm silver nanoparticles and Figure 2c for the two coupled particles as a function of the incident wavelength. The cross sections are normalized by the incident wavevector $k = 2\pi/\lambda$. Figure 1a and Figure 1ac show similar smooth increasing plots that does not exhibits local maxima. The absence of cavity resonance (also called Whispering Gallery Modes) is due to a weak refractive contrast between the microsphere and the surrounding background. On the contrary, the extinction cross section of the nanoparticle exhibits a peak due to a dipolar plasmon resonance occurring at $\lambda = 500$ nm. However, one can notice that the extinction cross section of the microparticle is much larger than the isolated particle even at the plasmon resonance. The addition of a nanoparticle in the photonic jet beaming does not increase the electromagnetic interactions with the incident field. The electromagnetic cross section of the system microsphere-particle is dominated by the microsphere. Consequently, the cross section of the nanoparticle has been increased.

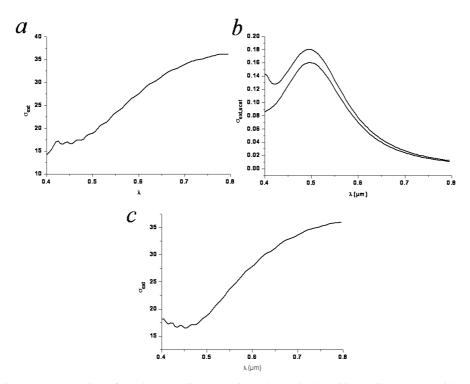


Figure 3. Extinction cross section of (a) the 2μ m diameter microsphere, (b) the 100 nm silver nanoparticle and (c) the two spheres system where the nanoparticles is located in the photonic jet produced by the microsphere at a distance d = 100 nm from the surface.

3.1 Backscattering

Figure 4a displays the power backscattered by the nanoparticles when located within the photonic jet and Figure 4c the backscattered power when the nanoparticle is located in the confined field produced by a Gaussian illumination on the microsphere. The backscattered power is obtained by integrating the Poynting vector of the scattered field in the far field over the half space z<0.

To estimate the benefits to employ a microsphere to focus light, the backscattered power plots have been normalized by the backscattered power when the nanoparticle is illuminated by (a) a plane wave and (c) a Gaussian beam. Figure 4a and 4c show that the microsphere enables to enhance the backscattered signal by an order of magnitude. However, it must be stressed that only 10% of the field scattered propagates in the backward direction. One can also notice that the backscattered power oscillates according to the relative positioning of the microsphere with the nanoparticle. This point will be interpreted later.

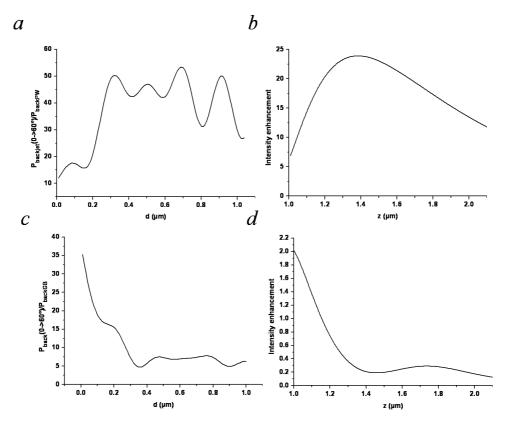


Figure 4: Backscattering enhancement of radiated power by a silver 100nm particle located at a distance d from the microsphere surface. (a) The microsphere is illuminated by an optical plane wave (b) the microsphere is illuminated by a Focused Gaussian beam. The power is normalized by the backscattered power when the same 100 nm nanoparticle is illuminated without microsphere. (b), (d) Intensity distribution along the z-axis for the same condition than in Figure 1a and 1c respectively.

Figure 4b and 4d displays the intensity enhancements along the propagating axis (z-axis) for conditions detailed in Figure 1a and Figure 4b. Comparison with Figure 4a and 4c underlines that if the oscillations are neglected, the backscattered power follow the same trend than the excitation field perceived by the nanoparticle. The backscattered power enhancement appears to be mainly due to the enhancement of the excitation field. One can then wonder where lies the benefit to employ photonic jets to detect metallic nanoparticles instead of far field focused beams? An answer can be find in the fact that microsphere are near field operating lenses. It has been demonstrated that evanescent components were weakly contributing to the field [13]. However, near-field operating devices have the strong advantage to modify radiation pattern of dipolar emitter located in their vicinity [24, 29-31].

4. DIPOLAR EMISSION CLOSE TO THE MICROSPHERE

4.1 Electric field components

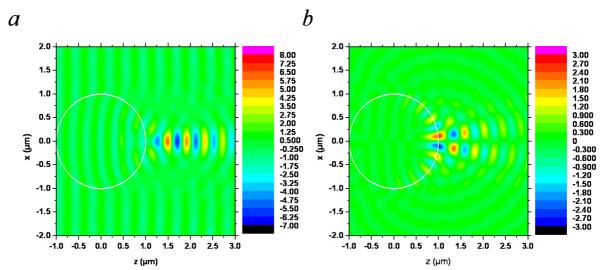


Figure 5: Electric field colored map (a) oriented along the incident polarization axis (i.e. x-axis) and (b) oriented along the propagation axis (i.e. z-axis) of a photonic beaming produced in the same conditions than Figure 1a.

The electromagnetic response of the nanoparticle depends on the features of the electric field excitation. The electric field amplitude along two directions of interest has been displayed in Figure 2. Figure 2a shows the total electric field polarized along the x-axis (incident polarization is along the x-axis) and Figure 2b displays the electric field that is directed along the z-axis. It clearly highlights that on the z-axis, the electric field is oriented along the x-axis while no electric field is oriented along the z-axis. The excitation field perceived by a nanoparticle located on the propagating axis of photonic jet beam is mainly along the polarization direction. As a consequence, the polarization moment induced in the particle as well as the field scattered will be polarized along the incident polarization.

A recent investigation performed by Agio et al. [32] has demonstrated that the high k-vectors constituting a focused beam tend to increase the dipolar behaviour of the particle, even for large nanoparticles. Although the size of the nanoparticle (100 nm) can enable a quadrupolar-type plasmon resonance in the optical range, for the considered frequency corresponding to a wavelength $\lambda = 500$ nm in vacuum, the particle mainly exhibits a dipolar resonance.

4.2 Emission of a dipole close to the dielectric particle

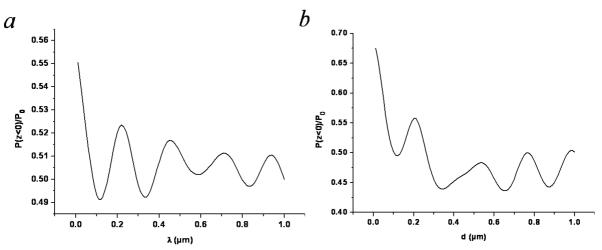


Figure 6: Power radiated in the half space z<0 when a dipole emitting at $\lambda = 500$ nm is located at a distance d from the microsphere of refractive index (a) $n_s = 1.6$, (b) $n_s = 2.44$.

Due to the physical considerations discussed above, the electromagnetic response of the nanoparticle is approximated by a dipolar emitter oriented along the x-axis (i. e. directed tangentially to the microsphere surface). The power radiated in the half space z<0 is displayed in Figure 6a as a function of the distance *d* between the sphere surface and the dipole. Oscillations appear in the radiated signal when a dipolar emitter is moved away from to the microsphere along the z-axis. The period of these oscillations is on the order of $\lambda/2$ which demonstrates the interference process occurring between the field radiated by the dipole and the field scattered by the microsphere. These interferences explain the oscillations observed in backscattered signal in Figure #. These oscillations enable an estimation of the positioning of a dipole close to a microsphere. Furthermore, Figure 6a clearly demonstrates that the microsphere favors the emission of the dipolar source in the z < 0 half space since more than 50% of the emitted power propagates in the z < 0 half space. It is even more conspicuous when the dipole is located closer than 100 nm from the microsphere.

Figure 6b shows the same plots that Figure 6a but for a high refractive index microsphere ($n_s = 2.44$). Figure 6b shows that the radiated power in the backward direction is enhanced when the dipole is located closer that 200 nm from the microsphere surface. It is yet counter intuitive to notice that if the dipole is located further than 200 nm from the microsphere surface, the field is principally radiated in the half space z>0.

The investigation of the backscattered power by a nanoparticle located in different beamings has demonstrated that a dielectric microsphere acting as a microlens can favor backscattering radiation. The enhancement of the backscattered signal has been investigated according to two contributions: the excitation field and the nanoparticle response. First, the microsphere enables to focus light and increase the excitation field on the nanoparticles by one order of magnitude. Then, the microsphere, operating in near field, can efficiently channel the optical response of a nanoparticle located closer than $\lambda/2$ from its surface, producing a redirection of the field scattered. In addition, the presence of the microsphere in the near field of the particle produces interferences responsible for oscillations in the backscattered power enabling estimation of the particle location at $\lambda/2$ scale.

It must be emphasized that a focused beam illumination on the microsphere enables to limit the longitudinal extent of the beam emerging from the sphere. Thus, this configuration enables a straightforward positioning of the nanoparticle at $\lambda/2$ scale since both the excitation field volume and the efficitive collection zone are coincident.

REFERENCES

- H. Rigneault, J. Capoulade, J. Dintinger, J. Wenger, N. Bonod, E. Popov, T. W. Ebbesen, P. F. Lenne, "Enhancement of single-molecule fluorescence detection in subwavelength apertures," Phys. Rev. Lett. 96, 117401 (2005).
- [2] E. Popov, M. Nevière, J. Wenger, P. F. Lenne, H. Rigneault, P. Chaumet, N. Bonod, J. Dintinger, T. W. Ebbesen, "Field enhancement in single subwavelength apertures," J. Opt. Soc. Am. A 9, 2342–2347 (2006).
- [3] H. Tamaru, H. Kuwata, H. T. Miyazaki, K. Miyano, "Resonant light scattering from individual Ag nanoparticles and particle pairs," Appl. Phys. Lett. **80**, 1826 (2002).
- [4] W. Rechberger, A. Hohenau, A. Leitner, J. R. Krenn, B. Lamprecht, F. R. Aussenegg, "Optical properties of two interacting gold nanoparticles," Opt. Comm. 220, 137–141 (2003).
- [5] K. Li, M. I. Stockman, D. J. Bergman, "Self-similar chain of metal nanospheres as an efficient nanolens," Phys. Rev. Lett. 91, 227402 (2003).
- [6] S. Bidault, F. J. García de Abajo, A. Polman, "Plasmon-based nanolenses assembled on a well-defined DNA template," J. Am. Chem. Soc. 130, 2750–2751 (2008).
- [7] Z. Chen, A. Taflove, V. Backman, "Photonic nanojet enhancement of backscattering of light by nanoparticles: A potential novel visible-light ultramicroscopy technique," Opt. Express **12**, 1214 - 1220 (2004).
- [8] X. Li, Z. Chen, A. Taflove, V. Backman, "Optical analysis of nanoparticles via enhanced backscattering facilitated by 3-D photonic nanojets," Opt. Express 13, 526 533 (2005).
- [9] S. Lecler, Y. Takakura, and P. Meyrueis, "Properties of a 3D photonic jet," Opt. Lett. 30, 2641-2643 (2005).
- [10] A. V. Itagi and W. A. Challener, "Optics of photonic nanojets," J. Opt. Soc. Am. A 22, 2847-2858 (2005).
- [11] J. Kofler, N. Arnold, "Axially symmetric focusing as a cuspoid diffraction catastrophe: scalar and vector cases and comparison with the theory of Mie," Phys. Rev. B. **73**, 235401 (2006).
- [12] P. Ferrand, J. Wenger, A. Devilez, M. Pianta, B. Stout, N. Bonod, E. Popov, and H. Rigneault, "Direct imaging of photonic nanojets," Opt. Express 16, 6930-6940 (2008).
- [13] A. Devilez, B. Stout, N. Bonod, E. Popov, "Spectral analysis of three-dimensional photonic jets," Opt. Express 16, 14200 – 14212 (2008).
- [14] M. Mosbacher, H.-J. Münzer, J. Zimmermann, J. Solis, J. Boneberg, P. Leiderer, "Optical field enhancement effects in laser-assisted particle removal," Appl. Phys. A: Mater. Sci. Process. 72, 41-44 (2001).
- [15] B. S. Luk'yanchuk, N. Arnold, S. M. Huang, Z. B. Wang, and M. H. Hong, "Three-dimensional effects in dry laser cleaning," Appl. Phys. A: Mater. Sci. Process. 77, 209-215 (2003).
- [16] A. Pereira, D. Grojo, M. Chaker, P. Delaporte, D. Guay, M. Sentis, "Laser-fabricated porous alumina membranes for the preparation of metal nanodot arrays," Small **4**, 572-576 (2008).
- [17] K. Piglmayer, R. Denk, and D. Bäuerle, "Laser-induced surface patterning by means of microspheres," Appl. Phys. Lett. 80, 4693-4695 (2002).
- [18] E. McLeod, C. B. Arnold, "Subwavelength direct-write nanopatterning using optically trapped microspheres," Nature nanotechnology 3, 413 - 417 (2008).
- [19] K. J. Yi, H. Wang, Y. F. Lu, Z. Y. Yang, "Enhanced Raman scattering by self-assembled silica spherical microparticles," J. Appl. Phys. 101, 063528 (2007).
- [20] J. Kasim, Y. Ting, Y. Y. Meng, L. J. Ping, A. See, L. L. Jong, S. Z. Xiang, "Near-field Raman imaging using optically trapped dielectric microsphere," Opt. Express 16, 7976 – 7984 (2008).
- [21] S.-C. Kong, A. Sahakian, A. Taflove, V. Backman, "Photonic nanojet-enabled optical data storage," Opt. Express 16, 13713 – 13719 (2008).
- [22] D. Gérard, J. Wenger, A. Devilez, D. Gachet, B. Stout, N. Bonod, E. Popov, H. Rigneault, "Strong electromagnetic confinement near dielectric microspheres to enhance single-molecule fluorescence," Opt. Express 16, 15297 – 15303 (2008).
- [23] A. Devilez, N. Bonod, J. Wenger, D. Gérard, B. Stout, H. Rigneault, E. Popov, "Three-dimensional subwavelength confinement of light using dielectric microspheres," Opt. Express 17, 2089 – 2094 (2009).
- [24] D. Gérard, A. Devilez, H. Aouani, B. Stout, N. Bonod, J. Wenger, E. Popov, H. Rigneault, "Efficient excitation and collection of single molecule fluorescence close to a dielectric microspheres," J. Opt. Soc. Am. B. 26, 1473, (2009).

- [25] A. Devilez, J. Wenger, B. Stout, N. Bonod, "Transverse and longitudinal confinement of photonic nanojet by compound dielectric microspheres," Proc. SPIE 7393, 73930E (2009).
- [26] B. Stout, M. Nevière, E. Popov, "Light diffraction by three-dimensional object: differential theory," J. Opt. Soc. Am. A 22, 2385 - 2404 (2005).
- [27] B. Stout, J.-C. Auger, J. Lafait, "A transfert matrix approach to local field calculations in multiple-scattering problems," J. Mod. Opt. 49, 2129 - 2152 (2002).
- [28] L. W. Davis, "Theory of electromagnetic beams," Phys. Rev. A 19, 1177 1179 (1978).
- [29] B. J. Soller, H. R. Stuart, D. G. Hall, "Energy transfer at optical frequencies to silicon-on-insulator structures," Opt. Lett. **26**, 1421, (2001).
- [30] J. J. Schwartz, S. Stavrakis, S. R. Quake, "Colloidal lenses allow high-temperature single-molecule imaging and improve fluorophore photostability," Nature nanotech. **5**, 127–132, (2009).
- [31] G. Pellegrini, G. Mattei, P. Mazzoldi, "Light extraction with dielectric nanoantenna arrays," ACS Nano 3, 2715–2721, (2009).
- [32] N. M. Mojarad, M. Agio, "Tailoring the excitation of localized surface plasmon-polariton resonances by focusing radially-polarized beams," Opt. Express. **17**, 117-122 (2008).