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Optical field enhancement effects on sapphire particles for different wavelengths and substrate media

R Zakaria, K S Hamdan, M I M Abdul Khudus and R Penny

Photonics Research Center, Faculty of Science, University of Malaya, 50603 Kuala Lumpur, Malaysia

E-mail: rozalina@um.edu.my

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Abstract
This paper presents a study on laser intensity distributions where field enhancements occur due to scattering by spherical sapphire particles and under conditions of different wavelengths and substrate media. A sapphire micro particle concept is employed to determine the effects of light–particle interaction with a particle refractive index approaching 2 (n ≤ 2). Mie scattering software is used to perform the computations for the presented results. Near-field effects are a consequence of irradiation scattering that occurs from spherical particles, and these effects play a fundamental role in nano-patterning and in diverse laser cleaning environments. Efficient laser cleaning requires the optimization of many parameters, and these simulated results involving sapphire particles at different laser wavelengths and in different substrate media allow for more judicious experimental implementations.

Keywords: near-field effect, particles, laser

1. Introduction
Near-field patterning was developed following studies by many researchers on the near-field effects of small particles on a substrate when subjected to laser irradiation [1–4]. A popular use of near-field enhancement is in the field of micromachining, whereby nonlinear absorption of the enhanced optical field between the spheres and substrate can contribute to the creation of nano-features on the substrate and open up a plethora of applications. This method of patterning has given rise to a number of interesting applications in optical devices and optical communication. One such example is doping with a soda-lime glass compound with consequent applications in medical diagnostics, laser glasses, undersea optical communication, integrated optical devices and optical data storage [5, 6]; it also has great potential for biochemical sensor applications using metallic nanostructures [7, 8] and plasmonic micro fabrication studies [9, 10].

Surface nano-patterning can be fabricated by numerous conventional processing techniques such as e-beam, x-ray and UV photolithography, including the near-field scanning optical microscope (NSOM) technique [11] and the laser-assisted atomic force microscopy (AFM/STM) tip patterning technique [12]. Research on laser-assisted surface patterning using micro particles, however, has focused primarily on particle-induced damage on the irradiated surface during dry laser cleaning using monodisperse silica SiO₂ [3, 13], polystyrene [7] or gold in a liquid environment [14]. Depending on the particle size and wavelength of the laser source, the optical field enhancement is due to either a lens focusing effect or Mie scattering by the spherical transparent dielectric particles [15]. This paper describes a Mie simulation study of near-field effects due to the scattering of laser beams by spherical sapphire particles in order to obtain optimal efficiency in dry or wet laser cleaning applications. The study considers different laser wavelengths and substrate media to allow the use of short or long laser wavelengths for efficient particle removal by means of laser cleaning.
Figure 1. Distribution of laser intensity \( I = IEI^2 \) at different laser wavelengths where maximum enhancement is about 110 times the illumination by a laser pulse of \( \lambda = 532 \) nm.

2. Theory

Spherical particles can act as spherical lenses [7], and as such are able to increase laser intensity if their diameter is greater than the laser wavelength. Light scattering in small particles has two limiting cases: the geometric optics limit (radius \( a \gg \lambda \)) and the electrostatic limit. The former limit is considered for the purposes of this study to allow the focusing of light rays in the particle. Optical intensity due to the focusing effect of the particle can be crudely estimated via the ray trace method of the particle’s caustics, utilizing Snell’s law and the principle of energy conservation [16].

In the geometric optics limit, spot size due to the focusing effect of a beam in a spherical particle can be approximated by the caustic of the particle as follows [17]:

\[
\omega \approx a \sqrt{\frac{(4 - n^2)^3}{27n^4}}
\]  

(1)

where \( a \) is the particle radius and \( n \) the refractive index, with the condition \( 1 < n < 2 \). A comparison with the numerical investigation based on the Mie formula [17] determines the spot size choice of

\[
\omega = \frac{\lambda}{5}
\]  

(2)

Furthermore, for sapphire \( n \) is close to 2 [18] independent of wavelength, which enhances the intensity of the optical properties in contrast to spheres with \( 1.1 < n < 1.8 \) that are weakly absorbing [17]. Large particles have stronger focusing effects that lead to further consideration of the temperature distribution and threshold fluence on the substrate. The investigation here is limited to the enhancement of an electric field due to the focusing effect of a particle which can be approximated as [19]

\[
\frac{I_m}{I_0} \approx \left( 1 + \frac{n^2 - 1}{n^2 + 2q^2} \right)^2
\]  

(3)

where \( I_m/I_0 \) represents optical enhancement and \( q = 2\pi a/\lambda \).

Depending on the particle size, intensities in the near-field of the particles can exceed the incoming laser intensity by considerably more than an order of magnitude. Such intensities cause local melting, ablation and plasma formation in the optical near-field of the particles [20]. Mie theory describes particle diameters that are comparable to or below the applied laser wavelength, and the associated size parameter is

\[
x = \frac{\pi d}{\lambda}
\]  

(4)

with \( d \) the particle diameter and \( \lambda \) the laser wavelength. Particular values of this size parameter result in a resonant intensity enhancement. This theory can be applied for spherical particles in homogeneous surroundings where the influence of the substrate can be neglected.

3. Results and discussion

Laser cleaning experiments tend to reveal damage to the removed particles [20, 21] or even nano-fabrication occurring on the substrate, and many parameters must be considered for efficient laser cleaning. Simulations that model Mie’s theory on the optical near-field region around a single particle have facilitated an understanding of the basic physics of the near-field focusing effect induced by a spherical particle. Calculations in all cases were performed for a free-space spherical particle with light interaction at the near-field region. Figure 1 shows results of an investigation of the near-field distribution for different laser illumination wavelengths (248, 355, 532 and 800 nm) in air for various particle sizes corresponding to \( a = 5\lambda \). The laser intensity distribution is a maximum at 532 nm \( (n = 1.77) \), which corresponds to approximately 110 times the source intensity, followed by 355 nm \( (n = 1.79) \), 800 nm \( (n = 1.76) \) and 248 nm laser \( (n = 1.84) \). Irradiation enhancement using a 532 nm laser on sapphire particles in air is shown in figure 2 to obtain a peak of almost 300 times the source intensity at the spherical inner
Figure 3. Schematic diagram of the three general cases: normal incidence and p- and s-polarized incident irradiation at different angles $\theta$ to a particle on the substrate.

region with a sharp decay outside the surface boundary of the sphere. The inset of figure 2 visualizes the highest intensity occurring within the sphere, which makes it inappropriate to use this wavelength for this particle. A lower intensity at the outer surface has the consequence of a low energy distribution consistently across the substrate surface. Optimum efficiency for the experimental process necessitates a careful selection of wavelength for a specific particle size and refractive index.

Numerous reports exist of the capabilities of KrF at $\lambda = 248$ nm [3, 22] and femtosecond laser at $\lambda = 800$ nm and 100 fs specification [23, 24]. These two wavelengths were considered in this work as comparisons for future experimental work. Future laser cleaning process studies will have a focusing particle placed on top of the substrate, and comparisons can be made with the theoretical predictions presented here. The maximum point is required to be as close as possible to the particle edge, and so $z/a = 1.1$ in this case. Studies of particles within media of different refractive indices were also undertaken to aid determination of the best environment for removal of particles, and media tested include air ($n = 1.0$), water ($n = 1.3$) and KOH liquid ($n = 1.409$). A selection of $n = 1.2$ shows the best conformance to the desired optimal of $z/a = 1.1$.

Figure 3 shows a schematic diagram for a particle on a substrate with the effects of p- and s-polarization of the incident radiation. The simulation in this paper has been performed for a particle at selected irradiation wavelengths while influence of the substrate is neglected, and so polarization does not play a role in these studies.

Figure 4(a) shows local field enhancement of the particle at 248 nm wavelength laser irradiation and a pulse duration of 23 ns. These Mie modeling results show a cross-sectional view of the normalized local field enhancement, $IEI^2$, underneath a single sapphire particle of 1.24 $\mu$m diameter and $n = 1.84$ in various media of differing refractive indices, under $x$-polarized plane wave excitation. Sapphire enhancement in air shows the highest intensity inside the particle, at around 160 times source intensity, and decays rapidly towards the edge. The enhancement field is associated with optical cavity resonance inside transparent particles; this can be considered as the interference of the incident beam and evanescent waves present in the vicinity of the particles [25] in the optical near-field region. Input wavelengths for this investigation are 248 and 800 nm, corresponding to particle diameters of 1.24 and 4 $\mu$m respectively. These values are chosen for the purposes of comparison in view of future experiments exploiting the accessibility of these two wavelengths. The focus point for a sapphire particle in water can be observed at position $z/a = 1.1$, which is very close to the particle surface. A further observation is the near-field enhancement decaying to half the maximum value in the range $z/a = 1.1$ to $z/a \sim 1.6$, which means that the particle surface must remain in the near-field distance for efficient patterning.

Figure 4. (a) Comparison of the intensity distribution in media with different refractive index ($n$) using an input illumination of 248 nm. (b) Light intensity $IEI^2$ distribution inside and outside 1.24 $\mu$m sapphire particles in water, with $n_{\text{water}} = 1.3$ and $n_{\text{sapphire}} = 1.84$. 

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Figure 5. (a) Comparison of the intensity distribution in media of different refractive index \( n \) using an input illumination of 800 nm. (b) Distribution of light intensity \( IEI^2 \) inside and outside 4 \( \mu \)m sapphire particles in water, with \( n_{\text{water}} = 1.3 \) and \( n_{\text{sapphire}} = 1.76 \).

Figure 4(b) gives the laser light intensity distribution calculated for this theory with an optical resonance effect in the near-field. Theoretical calculation using Mie theory proves that light energy is concentrated in a small region of less than 100 nm indicated by the red color region under the bottom of the particle [26]. Excimer laser irradiation at a wavelength of 248 nm on this sapphire particle causes laser light to be enhanced up to 60 times beneath the particle surface in the water environment. Light enhancement is also shown to increase when the particle diameter increases or the light wavelength decreases [27].

Near-field enhancement within the \( z/a \) range from 1.1 to 2.2 is seen in figure 5(a) to decay almost exponentially for this particle when immersed in water and KOH liquid and irradiated by 800 nm wavelength radiation. The focusing properties are observed to change depending on the surrounding medium. Enhancement also decreases from approximately 80 times in air to 60 times in water, and about 58 times in KOH liquid. In this latter case, the field enhancement decays from the focus point very slowly in comparison to the case in air, which results in a significant increase in the depth of focus [25] as shown in figure 5(b). This configuration is beneficial for laser processing of materials since the depth of focus extends the involvement of other factors such as the presence of neighboring particles, and facilitates fabrication of higher-aspect-ratio structures into a substrate.

Scanning electron microscopy (SEM) provides a top-view of \( \text{SiO}_2 \) in figure 6, allowing a comparison of sapphire with a \( \text{SiO}_2 \) particle of size 1.24 \( \mu \)m using a shorter wavelength of 248 nm. Figure 7 depicts the light intensity distribution with laser irradiation of wavelength 248 nm, whereby light intensity is enhanced up to 140 times in air under the \( \text{SiO}_2 \) particle (\( z/a = 1.0 \)) and decays quickly at \( z/a = 2.0 \). Light enhancement using this particle in other surrounding media is smaller; 60 times in water and 40 times in KOH liquid as the broadened peak indicates increasing depth of focus on the substrate.

4. Conclusion

Several conclusions can be made following this investigation. The likelihood of particle removal is affected by laser wavelength, as the wavelength determines the amount of thermal energy absorbed by the particle and/or substrate, or absorbed in other environments if wet or steam laser cleaning is used. Laser irradiation invoking a near-field effect is one of the effective ways to overcome the optical diffraction limit for surface nano-patterning applications. Sapphire particles here are assumed to have a spherical shape and effects of the substrate are neglected; examining the effects of using other substrates including polymer and arbitrary shapes of sapphire will be very interesting. Calculations here show that light enhancement increases as the laser wavelength decreases with respect to different surrounding media, and theoretical measurements such as threshold fluence can be considered.
Figure 7. (a) Comparison of the intensity distribution in media with different refractive index \( n \) using a SiO\(_2\) input illumination of 248 nm. (b) Light intensity \( I(E) \) distribution inside and outside a 1.24 \( \mu \)m particle immersed in air, where \( n_{air} = 1.6 \).

...as more significant contributing factors. An increased particle size results in a larger light enhancement area and consequent lower resolution for structure fabrications, which is undesirable for nanostructuring processes. The use of a 157 nm laser is feasible for high resolution structuring on the substrate [9, 28] although this wavelength is unfortunately easily absorbed in air and most optical materials.

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