Numerical investigation of passive optical sorting of plasmon nanoparticles

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Abstract: We explore the passive optical sorting of plasmon nanoparticles and investigate the optimal wavelength and optimal beam shape of incident field. The condition for optimal wavelength is found by maximising the nanoparticle separation whilst minimising the temperature increase in the system. We then use the force optical eigenmode (FOEi) method to find the beam shape of incident electromagnetic field, maximising the force difference between plasmon nanoparticles. The maximum force difference is found with respect to the whole sorting region. The combination of wavelength and beam shape study is demonstrated for a specific case of gold nanoparticles of radius 40nm and 50nm respectively. The optimum wavelength for this particular situation is found to be above 700nm. The optimum beam shape depends upon the size of sorting region and ranges from plane-wave illumination for infinite sorting region to a field maximising gradient force difference in a single point.

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References and links
1. Introduction

The preparation of highly monodispersed colloidal solutions of plasmon nanoparticles is crucial for any application where a narrow size distribution is required particularly to exploit their plasmonic properties. This includes biomolecular sensors [1], localised heaters [2] and photothermal imaging [3]. As most of these applications require colloids prepared in sterile way, the application of optical sorting offers an ideal solution not only because of the non-contact nature of sorting but also because of the exceptional sensitivity on particle size, shape and refractive index. Two main optical sorting approaches exist—namely active and passive. Active sorting techniques use fluorescent signals [4], optical switches [5, 6] and real-time computation [7, 8] for particle recognition that then subsequently acts as a trigger for another part of the system where optical (or other) forces are used to separate particles of different properties into separate streams. However, the need to use a trigger may add complexity and be impractical in many instances. For this reason, the last decade has seen the emergence of passive sorting methods which rely entirely on the different physical response of various particles to an extended optical field, commonly referred to as an optical potential energy landscape. Such passive sorting offers exceptional size and refractive index sensitivity [9, 10] and has been demonstrated in a number of geometries both with and without the presence of microfluidic flow. Sorting of particles in closed chambers with static fluid has been realised both by means of moving interference pattern [11, 12] or Bessel modes [13]. However, in the majority of sorting applications, laminar fluid flow perpendicular to the optical forces is employed and the separation of particles is either
realised solely by scattering force differences [14] in aperiodic optical patterns or by means of structured light fields [10, 15–19] creating periodical optical potential energy landscapes. In all these methods, the determination of the precise form of the applied optimal field, such that the maximum sorting sensitivity on size, shape and refractive index is achieved, remains an open question. To date it is also to be noted that passive optical sorting has been mainly considered solely with respect to micron-sized dielectric particles and cellular media. It is intriguing to consider how passive optical sorting may be extended to the domain of plasmon nanoparticles.

Here, we study passive optical sorting of plasmon nanoparticles and present a general two step approach that can be used to design the optimal illumination for sorting plasmon nanoparticles. In the first step, we find the optimal wavelength of illumination such that the separation is achieved with minimum temperature increase in the system. This is an important consideration in plasmonic systems as the excessive heat increases diffusion and convective effects, which is counter-productive in any sorting application. In the second step, the optimisation of illumination shape is realised using our force optical eigenmode (FOEi) [20] method, which can be readily applied to determine the optimal laser illumination field for sorting. The approach extends previous studies of forces on plasmon nanoparticles [21, 22] by considering the shape of the field, its wavelength and heating in the system at the same time. This paper is divided into three sections. In the first section, we present the FOEi method and derive formulae leading to optimised beam shape for sorting. The second part focuses on exploiting the plasmon resonances in the system and finding the optimal wavelength for sorting. The third part uses the optimal wavelength as input and extends the FOEi method to optimise the force difference over the whole sorting region.

2. Description of FOEi method

Our aim is to optimise the incident electromagnetic field $E_{inc}$ in a way that maximises the exerted force $F^{\mu} = F \cdot u$ in a specified direction $u$ (Fig. 1). Since our incident field of angular frequency $\omega$ can be decomposed into a sum of $\mu$ monochromatic plane waves ($e^{i\omega t}$), we can write (using summation over repeating indices)

$$E_{inc} = a^{\mu} E_{inc}^{\mu},$$

where $a^{\mu}$ are the complex expansion coefficients and $E_{inc}^{\mu}$ are the incident plane waves. The $a^{\mu}$

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{fig1.png}
\caption{We look for an amplitude and phase $a$ of a given set of plane waves such that the force $F^{(1,0)}$ is maximised.}
\end{figure}
coefficients modulate both phase and amplitude of each and single plane wave independently. The scattered field generated upon interaction with the particle has the same expansion coefficients, so that we can write \( E_{sca} = d^\mu E_{inc}^\mu \). The final field \( E \), which is a sum of incident and scattered field, can then be written as

\[
E = d^\mu (E_{inc}^\mu + E_{sca}^\mu) = d^\mu E^\mu,
\]

where \( E^\mu \) is a solution of the scattering problem for an incident field given by plane wave \( E_{inc}^\mu \).

The optical-cycle averaged electromagnetic force in the direction \( u \) is given by

\[
F^u = \langle F \rangle u_t = \oint_C \langle \sigma_{ij} \rangle n_j u_i \, ds,
\]

where \( n_j \) is outward unit normal to an element \( ds \) of the curve \( C \) enclosing the particle for which we optimise the force and \( \langle \cdot \rangle \) is optical-cycle average. The Maxwell stress tensor, \( \sigma_{ij} \), can be written for the final field \( E \) as [23]

\[
\langle \sigma_{ij} \rangle = \frac{1}{4} \left[ \varepsilon_0 \varepsilon_m (d^\mu E^\mu)^* (a^\nu E^\nu)_j + \mu_0 \mu_m (d^\mu H^\mu)^* (a^\nu H^\nu)_j \right. \\
- \varepsilon_0 \varepsilon_m (a^\nu E^\nu)_j (d^\mu E^\mu)^* + \mu_0 \mu_m (a^\nu H^\nu)_j (d^\mu H^\mu)^* \left. \right] - \delta_{ij} \left[ \varepsilon_0 \varepsilon_m (d^\mu E^\mu)^* (a^\nu E^\nu)_k + \mu_0 \mu_m (d^\mu H^\mu)^* (a^\nu H^\nu)_k \right].
\]

Using Eq. (2), we can rewrite Eq. (4) as

\[
\langle \sigma_{ij} \rangle = \langle d^\mu \rangle^* \left[ \frac{1}{4} \left[ \varepsilon_0 \varepsilon_m (d^\mu E^\mu)^* (a^\nu E^\nu)_j + \mu_0 \mu_m (d^\mu H^\mu)^* (a^\nu H^\nu)_j \right. \\
+ \varepsilon_0 \varepsilon_m (a^\nu E^\nu)_j (d^\mu E^\mu)^* + \mu_0 \mu_m (a^\nu H^\nu)_j (d^\mu H^\mu)^* \left. \right] - \delta_{ij} \left[ \varepsilon_0 \varepsilon_m (d^\mu E^\mu)^* (a^\nu E^\nu)_k + \mu_0 \mu_m (d^\mu H^\mu)^* (a^\nu H^\nu)_k \right] \right] a^\nu.
\]

and after rearrangement of the expansion coefficients \( d^\mu \) we obtain

\[
\langle \sigma_{ij} \rangle = \langle d^\mu \rangle^* \left[ \frac{1}{4} \left[ \varepsilon_0 \varepsilon_m (E^\mu)^* E^\nu_j + \mu_0 \mu_m (H^\mu)^* H^\nu_j + \varepsilon_0 \varepsilon_m (E^\mu)^* (E^\nu)_j + \mu_0 \mu_m (H^\mu)^* (H^\nu)_j \right. \\
+ \varepsilon_0 \varepsilon_m (E^\mu)^* (E^\nu)_k + \mu_0 \mu_m (H^\mu)^* (H^\nu)_k \left. \right] - \delta_{ij} \left[ \varepsilon_0 \varepsilon_m (E^\mu)^* E^\nu_k + \mu_0 \mu_m (H^\mu)^* (H^\nu)_k \right] \right] a^\nu.
\]

Substituting Eq. (6) into Eq. (3) gives

\[
F^u = \langle d^\mu \rangle^* \left[ \int_C \left[ \frac{1}{4} \left[ \varepsilon_0 \varepsilon_m (E^\mu)^* E^\nu_j + \mu_0 \mu_m (H^\mu)^* H^\nu_j + \varepsilon_0 \varepsilon_m (E^\mu)^* (E^\nu)_j + \mu_0 \mu_m (H^\mu)^* (H^\nu)_j \right. \\
+ \varepsilon_0 \varepsilon_m (E^\mu)^* (E^\nu)_k + \mu_0 \mu_m (H^\mu)^* (H^\nu)_k \left. \right] - \delta_{ij} \left[ \varepsilon_0 \varepsilon_m (E^\mu)^* E^\nu_k + \mu_0 \mu_m (H^\mu)^* (H^\nu)_k \right] \right] n_j u_i \, ds \right] a^\nu.
\]

or more simply in matrix form

\[
F^u = a^\dagger Ma,
\]

where the matrix coefficients \( M^{\mu\nu} \) are given by the line integrals in Eq. (7) and \( a \) is the vector form of \( d^\mu \). We remark that the matrix is Hermitian \( (M = M^\dagger) \) and thus its eigenvalues are real. This means that the force \( F^u \) is in a symmetric sesquilinear form, which is just an extension of quadratic form to complex numbers. Any symmetric sesquilinear form can be visualised as

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an ellipsoid with the length of principal axes equal to the eigenvalues $\lambda_n$ of the matrix $M$. This has far reaching implications for our optimisation process since the surface of the ellipsoid generated by the symmetric sesquilinear form is extremized at the end points of principal axes. This means that finding the eigenvalues $\lambda_n$ of the matrix $M$ and selecting the largest one from the set extremizes our problem. The eigenvector (force optical eigenmode) $a_{\text{max}}^n$ corresponding to maximum eigenvalue $\lambda_{\text{max}}^n$ (given by $M a_{\text{max}}^n = \lambda_{\text{max}}^n a_{\text{max}}^n$), then provides the necessary information about amplitude and phase of incident plane waves $E_{\text{inc}}^\mu$ in Eq. (1) so that the force is maximised. Experimentally, the optimised $a_{\text{max}}^n$ can be created using spatial light modulator (offering control of both phase and amplitude) in the system placed in conjugate plane [20] with respect to the back-focal plane of a microscope objective.

We remark that the above described method optimises the force on one type of particle at a single point only. However, the method can be easily extended to provide the optimised illumination for the force difference over the whole sorting region for two types of nanoparticles. This is discussed later in the paper.

**Numerical considerations:** We use COMSOL Multiphysics v4.1 RF module in scattering formulation to calculate the total field solutions $E^\mu$ for corresponding incident plane waves $E_{\text{inc}}^\mu$. We first find the solutions $E^\mu$ for particle $p_1$. We subsequently use the solutions $E^\mu$ to find the elements $M_{1\mu\nu}$ of matrix $M_1$ and determine the eigenvalues and corresponding eigenvectors. The principal eigenvector gives the optimum force for particle $p_1$ in a single position. The same procedure is repeated for a second type of particle, $p_2$, delivering matrix $M_2$. The matrices $M_1$ and $M_2$ then encode all the information about interactions of the incident fields with the particles. Finding the matrix elements $M_{\mu\nu}$ of matrix $M$ is computationally very intensive as combinations of $N(N+1)/2$ solutions need to be constructed and integrated over a sphere boundary. Here $N$ denotes number of plane waves in the angular spectrum representation and thus the number of pixels on spatial light modulator. As the azimuthal discretization of angular spectrum representation increases the number of combinations in 3D significantly, we have restricted the simulations to 2D to illustrate the method.

We can obtain educated estimates of 3D values from 2D values by extruding the 2D circle of radius $r$ by $d$ such that it creates a cylinder with a volume equal to the volume of the sphere with the same radius. The extrusion factor $d$ is given by

$$\pi r^2 d = \frac{4}{3} \pi r^3 \rightarrow d = \frac{4}{3} r.$$  \hspace{1cm} (9)

The validity of the COMSOL model was tested in 3D by comparing the optical forces and scattering and absorption efficiencies with Mie theory. The difference between the COMSOL model and the Mie model was less than 2 percent. In 2D, optical forces and scattering and absorption efficiencies were calculated using several independent methods to ensure the model validity.

3. **Plasmon resonances in the system**

We choose as our testing system gold nanoparticles [24] of radius $r_1 = 50\text{nm}$ and $r_2 = 40\text{nm}$. We consider a substrate of glass with refractive index $n_g = 1.5$. The particles are assumed to be dispersed in water with $n_w = 1.33$ (Fig. 1).

Plasmonic resonances offer exceptional sensitivity on size. However, for nanoparticles of very similar sizes, the force difference generated solely by the plasmon resonance in the system is still rather small. In case of dielectrics, increasing the intensity offers simple solution in such a situation, however plasmonic resonances are associated with non-negligible heat generation and as such increasing the intensity may produce increased diffusion rates and convective currents, which will interfere with sorting efforts. It is thus beneficial to find a wavelength for...
which the sorting effects are maximised and heating is minimised. Further, multiple laser wavelengths in a counter-propagating geometry with carefully adjusted powers can be employed to exploit the small plasmon resonance differences of nanoparticles. Our method can solve for optimum illumination for each and single wavelength separately; however, the use of multiple laser wavelengths is expensive and increases complexity of the system. It is therefore of interest to optimize the force differences using a single laser wavelength.

We choose the p-polarisation for the incident plane waves as the kind of plasmon resonance supported by the sphere appears in 2D for p-polarisation. The s-polarisation would only induce movement of electrons along the infinite cylinder. Figure 2(a) shows the scattering $Q_{\text{sca}}$ and absorption $Q_{\text{abs}}$ efficiencies for $r_1 = 50\text{nm}$ gold nanoparticle. Note that the 3D efficiencies calculated from Mie theory and the corresponding 2D efficiencies (transformed to 3D) for p-polarisation follow very similar pattern, which differs only in amplitude and a slight blue shift of 2D resonance peaks with respect to 3D resonances. Figure 2(b) shows the 3D forces along the substrate acting on the gold nanoparticles calculated for plane-wave incident at near critical angle of $\theta = 64^\circ$. The slight shift in resonances due to the different sizes of nanoparticles creates a force difference improvement with a peak around 550nm (black curve in Fig. 2(b)). If we do not take into account the proximity of the spheres to the surface (Faxen’s correction [25]) and assume we are in a low Reynolds number regime, then the drag force is given by the Stokes equation. Neglecting inertial effects we can equal optical and drag force and obtain the

Fig. 2. a) Scattering $Q_{\text{sca}}$ and absorption $Q_{\text{abs}}$ efficiencies calculated using Mie theory for 3D nanoparticle of $r_1 = 50\text{nm}$ and the corresponding 2D values converted to 3D equivalents. Nanoparticle is in water with $n_w = 1.33$; b) Forces and their respective difference $\Delta F$ acting on $r_1 = 50\text{nm}$ and $r_2 = 40\text{nm}$ gold nanoparticles. Forces are parallel to the substrate plane. The illumination is a plane-wave at near critical angle of $\theta = 64^\circ$ with power density corresponding to $1\text{mW/\mu m}^2$; c) Speed difference $\Delta v$ and temperature increase $\Delta T$ for the same illumination as in b); d) Speed difference normalised with respect to the temperature increase in the system.
expression for the particle speed in 3D as

$$v_{3D} = \frac{2F_{2D}}{9\pi \eta}, \quad (10)$$

where the force $F_{2D}$ was calculated in 2D using Eq. (8) and the dynamic viscosity of water at $T = 20{\degree}C$ is $\eta = 1.002 \times 10^{-3} \text{ Pa} \cdot \text{s}$. Figure 2(c) shows the speed difference generated by force differences in Fig. 2(b) and the average temperature increase as the particle enters the sorting field. The estimate of average temperature increase was calculated using [26] (neglecting particle movement and proximity of glass surface)

$$\Delta T = \frac{Q_1 + Q_2}{4\pi \kappa_0 \left(\frac{r_1^2 + r_2^2}{2}\right)}, \quad (11)$$

where $Q_1$ and $Q_2$ are 3D powers of heat generation in nanoparticles and $\kappa_0 = 0.6 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ is the thermal conductivity of water. This temperature increase is reached very quickly as the particle enters the sorting field. Results in Fig. 2(c) and Fig. 2(d) suggest, that heating at wavelengths above 700nm is quite low and the sorting speed remains high. For this reason we set the vacuum wavelength for our method to $\lambda_0 = 700$nm. Please note that a slight (30nm) blue shift in the resonances of 2D case does not have significant impact on the choice of this wavelength for the 3D scenario.

4. Optimising the force difference in region of interest (ROI)

Using the optimum wavelength from previous section we can proceed with optimisation of the beam shape. We introduce a 10nm separation between the lowest point of particle and interface, which closely mimics a typical experimental situation. The set of $N$ incident plane waves defined by $k_\theta$ vectors ($\theta = (-70^\circ, ..., 70^\circ)$ with a step of $2^\circ$) is used for discretization (see Fig. 1). The limits of $\theta$ correspond to experimental limitation for $NA = 1.4$ oil immersion objective. The goal is to optimise the force difference along $u = (1, 0)$.

Using matrices $M_1$ and $M_2$, the equation for force difference in a single point for our choice of particles is

$$\Delta F = F_1^u - F_2^u = a^\dagger (M_1^u - M_2^u) a = a^\dagger D_{12}^u a. \quad (12)$$

Due to the symmetry of the system two optimum solutions exists - one optimising the force difference in $+x$ direction and the second for $-x$ direction. In subsequent discussions we always choose the solution optimising the force in $+x$ direction. The field locally optimising the force difference for gold nanoparticles of our choice is on Fig. 3. Notice that the field creates a very strong field gradient in the $+x$ direction around point $x = 0$, where we want to maximise the force difference for our testing particles. Also notice that the back focal plane pattern corresponding to this field has significant contributions from plane waves propagating in the opposite $-x$ direction. Although this might seem surprising, we need to realise that the final goal of our method is to interfere the plane waves in such a way to create the strongest gradient in $+x$ direction. Apparently the counter-propagating waves increase the number of degrees of freedom for efficient interference leading to strong gradient and are thus utilised automatically by the FOEi method. It also make sense that the increased intensity at the back focal plane appears for near critical angle plane waves as those plane waves contribute the most to the intensity near the glass/water interface. Note that the phase at the back focal plane is also significantly altered to maximise the force difference.

So far our method optimises the force difference locally. To expand this approach to a larger region we need to optimise $\Delta F$ over a certain range, in our case line segment defined by $x =$
Fig. 3. Field optimising the force difference $\Delta F$ for gold nanoparticles of radius $r_1 = 50\text{nm}$ and $r_2 = 40\text{nm}$ in a single point $x = 0$ with corresponding amplitude $|a^\mu|$ and phase $\arg(a^\mu)$ for each plane wave from angular spectrum. The $|a^\mu|$ and $\arg(a^\mu)$ correspond to the pattern at the back focal plane of the objective. This illumination of the back focal plane forms a very strong field gradient in $+x$ direction in the focal plane, which maximises the force difference for our testing particles. Note that scattering from the particles is not included.

$\langle -l, l \rangle$. Displacing the particle in $x$-direction causes the particle to experience different relative phases between the fields $E^\mu$. Since we use combination of solutions to calculate matrix $M$ this relative phase can be taken into account using

$$M^{\mu\nu}(x) = e^{i k_w \sin(\theta^\mu) x} [M^{\mu\nu}] e^{-i k_w \sin(\theta^\nu) x}$$

(13)

where $k_w = (2\pi/\lambda_0)n_w$ and $\theta^\mu$ is the angle of the plane wave after the glass/water interface for each incident plane wave $E_{inc}^\mu$. $M^{\mu\nu}$ is the matrix calculated for particle at position $x = 0$. Using the translation relations for matrix $M^{\mu\nu}$ we obtain

$$\Delta F(l) = (a^\mu)^* \left[ \frac{1}{2l} \int_{-l}^l e^{ik_w x (\sin(\theta^\mu) - \sin(\theta^\nu))} \, dx \right] (M_1^{\mu\nu} - M_2^{\mu\nu}) a^\nu$$

$$= (a^\mu)^* \left[ \sin(k_w l (\sin(\theta^\mu) - \sin(\theta^\nu))) (M_1^{\mu\nu} - M_2^{\mu\nu}) \right] a^\nu$$

$$= a^\dagger R_{12}^u(l) a.$$  

(14)

The illumination optimisation is then performed on the modified matrix $R_{12}^u(l)$, which has the same input matrices $M_1^{\mu\nu}$ and $M_2^{\mu\nu}$ for all values of $l$. Finding the optimum illumination for a ROI of any size is thus very efficient.

Optimal illumination for ROI sizes of $l = 500\text{nm}$ (Fig. 4) and $l = 5\mu\text{m}$ (Fig. 5) differs significantly from the single point optimised problem (Fig. 3). The optimised field corresponds in its bulk to the focusing of light into ROI. The solution is quite close to the Gaussian beam send to the edge of the back focal plane of the objective. However, the phase for plane waves above critical angle is significantly modulated and the intensity profile is not entirely Gaussian. The width of the beam at the back-focal plane optimising the $l = 5\mu\text{m}$ situation is also noticeable.
Fig. 4. Field optimising the force difference $\Delta F$ for $l = 500\text{nm}$. The phase of $\phi^l$ is plotted in the region where it is well-defined. Notice that the field is focused into the ROI. The left edge of the back focal plane contributes the most to the optimised field in the focal plane. The phase at the back focal plane is slightly modulated as well.

Fig. 5. Field optimising the force difference $\Delta F$ for $l = 5\mu m$. The shape of the beam at the back-focal plane of the objective has a narrow distribution of amplitude in the proximity of critical angle.
smaller than for the case of \( l = 500\,\text{nm} \). This is a direct consequence of Eq. (14). As we increase \( l \), the off-diagonal terms in matrix \( \mathbf{R}_{12}(l) \) become less important due to the behaviour of the \( \text{sinc} \) function as \( l \) increases. In the limit \( l \to \infty \) only the diagonal terms remain. This means that the eigenmodes (eigenvectors of \( \mathbf{R}_{12}(l) \)) in this case correspond to single plane waves as defined in our initial set. The eigenmode (plane wave) with maximum eigenvalue optimises our problem for an infinitely large ROI. The solution found by the FOEi method for \( l = 100\,\text{mm} \)

Fig. 6. Field optimising the force difference \( \Delta F \) for \( l \to 100\,\text{mm} \). As the phase of \( a^\mu \) for zero amplitude \( |a^\mu| \) is not well defined, it is not displayed in the graph. The optimum angle plane wave is \( 64^\circ \), which is close to critical angle for given interface.

(Fig. 6) is the plane wave near the critical angle. As the phase of \( a^\mu \) for zero amplitude \( |a^\mu| \) is not well defined, it is not displayed in the graph. The result validates that the FOEi method is working correctly, as the near critical angle plane wave provides the highest intensity and force difference at the interface for infinite system.

The force difference is significantly increased in the cases of small ROI sizes compared with plane wave illuminated system that optimises the sorting for infinite sorting space (Fig. 7(a)). The bulk of this improvement is due to increased intensity of light in ROI, but the periodic pattern of forces indicates a more complex response of the system. This periodic pattern is not related to the discretization of \( k \)-space, where we expect periodicity to appear around \( 14\,\mu\text{m} \).

Naturally, we do not wish the sorting ROI to become too small as the experimental realisation would become increasingly complicated. It is interesting to look at the dependence of \( \Delta F \) on the size of ROI. To show the improvement compared to infinite system, we normalise \( \Delta F \) by \( \Delta F_{pw} \), where \( \Delta F_{pw} \) is the force difference for optimised infinite system (pw stands for plane wave). The result (Fig. 7(b)) indicates that the gain is significant for a wide range of experimentally interesting ROI sizes. The dip around \( l = 14\,\mu\text{m} \) is present due to discretization of \( k \)-space described above. The increase in ratio \( \Delta F / \Delta F_{pw} \) for \( l > 14\,\mu\text{m} \) is then equivalent to the formation of second beam focus in ROI due to onset of periodicity.

\textit{Discussion of results:} The FOEi method is capable of finding the optimal beam shape for illumination such that the force difference is maximised over the whole sorting region. The computationally intensive calculation of matrices \( \mathbf{M}_1 \) and \( \mathbf{M}_2 \) is compensated by the fact that
Fig. 7. (a) Blue and green points show forces acting on individual particles in optimised field for $l = 500$ nm. Red points show the force difference. The coloured lines show the same but for plane wave illumination optimising the infinite system. (b) The ratio $\Delta F/\Delta F_{pw}$ as a function of ROI size. The dip around $l = 14 \mu m$ is due to k-space discretization of angular spectrum representation. The gain in $\Delta F$ is significant in the experimentally interesting region.

the same matrices can be used for finding optimal illumination for any size of sorting region. We note that even though the solution does not optimise the vertical force pointing towards the substrate, we found that this is the case for all our solutions. However, the sign of this force is wavelength and particle size dependent and as such the attractive vertical force is not a general feature of the method. Further, the vertical force is not constant in the sorting region, which might introduce some modulation of force difference due to the Faxen correction. To resolve this one may minimise the vertical force and use an auxiliary beam with constant vertical force over the whole sorting region. This would restrict the diffusion of particles in vertical direction in more controlled way. It is possible to modify the method to simultaneously optimise for several parameters of the system. In our case, the full optimised solution for sorting applications of plasmon nanoparticles would involve simultaneous maximisation of force difference in ROI, minimisation of vertical force, and minimisation of heating. Such a problem reduces to finding matrices (operators) for all parameters of interest and choosing the eigenmodes optimising for such a set of parameters. It is very interesting, for plasmonic sorting in general, to find the beam shape of the field maximising the force difference and minimising the heating. This would also clearly identify the contribution of focusing to the overall improvement of force difference. This is a focus of our ongoing research.

5. Conclusion

We successfully optimised the illumination for sorting gold nanoparticles using our two step approach. Firstly, we found the optimal wavelength maximising the nanoparticle separation and minimising the temperature increase in the system. This is an important consideration in plasmonic systems as the excessive heat increases diffusion and convective effects. Secondly, we found the optimum beam shape of the illumination field for sorting using the method of force optical eigenmodes (FOEi). The applicability of the method was numerically demonstrated for the special case of sorting gold nanoparticles of different size. We plan to extend our approach and perform simultaneous optimisation of several parameters of interest for sorting applications, e.g., minimised heating, maximised force difference along substrate and minimised vertical force. This involves finding operators for all of parameters of interest and choosing the
eigenmodes optimising them. This will be subject of further work along with the efficient extension of the FOEi method to 3D case.

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