Design considerations of gold nanoantenna dimers for plasmomechanical transduction: supplement

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1. COMSOL MODEL VS MIE THEORY

![Graph showing Comparison of scattering spectra of 100 nm diameter gold nanosphere calculated with COMSOL multiphysics with Mie scattering calculated using Mie theory.]

**Fig. S1.** Comparison of scattering spectra of 100 nm diameter gold nanosphere calculated with COMSOL multiphysics with Mie scattering calculated using Mie theory.

Scattering properties of a 100 nm diameter gold nanosphere were calculated in the visible region using dielectric function from Johnson and Christy [1]. In Fig. S1 we can see a good agreement of the COMSOL model with scattering properties calculated using Mie theory.

2. SCATTERING SPECTRA OF NANO DISC DIMERS

The calculated scattering spectra of nanodisc dimers of radius 50 nm, 75 nm and 100 nm are shown in Fig. S2a, S2b and S2c respectively. A red-shift in the resonance peak is observed with decreasing inter-particle distances for all dimer cases. Such behavior is displayed mainly due to increased plasmonic coupling between the nanodiscs, shifting the plasmon resonances to lower energies, accompanied with broadening of the scattering spectra.

3. ELECTRIC FIELD ENHANCEMENT IN NANO DISC DIMERS

Electric field enhancement was calculated for nanodiscs of radii 50 nm, 75 nm and 100 nm at gap distances of 50 nm and 10 nm respectively. Light was incident perpendicular to the plane of nanodiscs with electric field aligned along the inter-particle axis of the dimer. Fig. S3a, S3c and S3e show the electric field enhancement for nanodisc dimers of radius 50, 75 and 100 nm with 50 nm separation distance. Similarly, electric field enhancements at an inter-particle distance of 10 nm for discs of radius 50 nm, 75 nm and 100 nm can be seen in Fig. S3b, S3d and S3f respectively. The excitation wavelength for the disc dimers was determined from their scattering spectra. For each case the excitation wavelength was chosen to be at the resonance conditions. We observe a strong electric field enhancement between the discs as the distance between them decreases from 50nm to 10nm. This occurs as a consequence of stronger plasmon coupling emerging at smaller inter-particle distances. The largest enhancement factor of 43 is observed for the 100 nm
Fig. S2. a Scattering cross-section for a gold nanodisc dimer of radius 50 nm at varying inter-particle distances 10–50 nm under plane wave excitation. b Scattering cross-section for a gold nanodisc dimer of radius 75 nm at varying inter-particle distances 10–50 nm under plane wave excitation. c Scattering cross-section for a gold nanodisc dimer of radius 100 nm at varying inter-particle distances 10–50 nm under plane wave excitation.
radius nanodisc dimer at a separation distance of 10 nm. The electric field enhancement for 50, 75 and 100 nm radius discs with 50 nm separation reveal a dipolar mode profile at resonance excitation conditions, with confinement regions mainly being present in the gap between the dimer and at the outer edges of the nanodisc. As the distance between nanodiscs decreases to 10 nm, the confinement in the gap region increases significantly, forming hot-spots for electric field enhancement.

4. POLARISATION DENSITY MAPS OF NANODISC DIMERS

The polarisation maps of nanodisc dimers were obtained by calculating the charge induced on the nanodisc dimers upon optical excitation. Incident light was injected along z-axis with electric-field vector aligned along the x-axis. Fig. S4a shows the charge distribution on the 50 nm radius nanodisc dimer and Fig. S4b on 100 nm radius nanodisc dimer, with 10 nm gap between the nanodiscs. The maps reveal emergence of opposite charge centers at the top and bottom faces of the nanodiscs, which is identical for nanodiscs of both radii. The larger disc dimer of radius 100 nm attains a larger magnitude of polarisation density \(20 \times 10^{-12} \text{ C/m}^2\) compared to \(4 \times 10^{-12} \text{ C/m}^2\) observed in smaller nanodisc dimer of radius 50 nm. This implies that a stronger electrostatic interaction exists between the nanodiscs of larger radius, leading to a stronger attractive force between them.

5. OPTICAL FORCE STUDIES IN NANODISC DIMERS USING GAUSSIAN BEAM EXCITATION

Fig. S5 shows the attractive optical forces in nanodisc dimers of radius 50, 75 and 100 nm respectively at varying inter-particle distances between 10 and 50 nm. Nanodisc dimers were excited using a Gaussian beam source where the excitation wavelength for each radius was chosen according to the plasmon resonance condition for the dimer at 10 nm gap between the discs. The 50 nm radius nanodiscs were excited at 625 nm, 75 nm radius nanodiscs at 745 nm and 100 nm nanodiscs at 900 nm respectively. The non-paraxial approximation was employed for generating the Gaussian beam, resulting in a beam width of 625 nm for exciting 50 nm nanodisc dimers, 745 nm beam width for exciting 75 nm radius nanodisc dimers and 900 nm beam width for exciting 100 nm radius nanodiscs. The magnitude of electric-field was 1 V/m for all cases of excitation.

6. EFFECT OF ROUNDING OF THE EDGES ON SCATTERING PROPERTIES OF NANOCUBE DIMER

We observe the effect of the degree of sharpness of the edges of a nanocube dimer on its optical properties in Fig. S6. A blue-shift in the scattering spectra of a nanocube dimer of edge length 75 nm and 10 nm gap distance is observed upon rounding its edges by a radius of 5 nm.

7. ELECTRIC FIELD ENHANCEMENT IN NANOCUBE DIMERS

Electric field (E-field) enhancement was calculated in nanocube dimer of edge length 100 nm and 150 nm at an inter-particle separation of 10 nm. The nanocube dimers were excited by plane wave travelling perpendicular to the plane of the cubes and the orientation of the electric field was kept along the inter-particle axis of the nanocubes for plasmonic coupling between the nanocubes. Fig. S7a, S7c and S7e show the E-field enhancement profile in nanocube dimers with 100 nm edge length at 604 nm, 670 nm and 800 nm respectively. At resonant excitation condition (670 nm), a strong localisation of the electric field is seen in the gap region between the nanocubes, also referred to as the hot-spot for local field enhancement. When the excitation source wavelength is red-shifted w.r.t. the plasmon resonance to 800 nm, the regions of localisation of the E-field shift position towards outer edges of the nanocubes. In case of excitation wavelength of 604 nm, which is blue-shifted to the resonance, the localisation can be seen present at outer corners of the nanocube dimer. Fig. S7b, S7d and S7f show the electric-field enhancement profile in nanocube dimer of edge-length 150 nm at 880 nm, 862 nm and 936 nm respectively. Similar to 100 nm edge length nanocube dimer, the localisation of the E-field at resonant excitation (862 nm) is observed in the gap region between the two nanocubes. On changing the excitation source wavelength to 880 nm, the localisation becomes asymmetric in the gap region. Upon red-shifting the excitation wavelength further to 936 nm, the localisation of the E-field shifted position towards the outer
**Fig. S3.** X-Z view of electric-field profile in gold nanodisc dimers under plane wave excitation normal to the plane of nanodiscs. 

- **a** 50 nm radius nanodisc dimer with 50 nm gap at 576 nm excitation.
- **b** 50 nm radius nanodisc dimer with 10 nm gap at 620 nm excitation.
- **c** 75 nm radius nanodisc dimer with 50 nm gap at 654 nm excitation.
- **d** 75 nm radius nanodisc dimer with 10 nm gap at 749 nm excitation.
- **e** 100 nm radius nanodisc dimer with 50 nm gap at 759 nm excitation.
- **f** 100 nm radius nanodisc dimer with 10 nm gap at 906 nm excitation.
Fig. S4. Polarisation density maps of gold nanodisc dimers under plane wave excitation. a Polarisation density map for a 50 nm radius nanodisc dimer with 10 nm gap distance at 620 nm excitation. b Polarisation density map for a 100 nm radius nanodisc dimer with 10 nm gap distance at 906 nm excitation.

Fig. S5. Optical forces in gold nanodisc dimers of diameter 100, 150 and 200 nm for Gaussian beam excitation. Nanodisc dimer with 100 nm diameter were excited at 625 nm, 150 nm diameter at 745 nm and 200 nm diameter at 900 nm respectively, with spot size equal to excitation wavelength for each case.
edges of the nano-dimer. Compared to nanocubes with 100 nm edge length, greater enhancement of E-field was observed in nanocubes with 150 nm edge length.

8. SPECTRAL RESPONSE OF OPTICAL FORCES IN NANODISC AND NANOBAR DIMERS

The relation of optical forces with wavelength was studied in nanodiscs and nanobars at an inter-particle distance of 10 nm. The scattering properties of the respective geometries were plotted to observe the correlation between the optical properties and the internal plasmonic forces present in them. The optical force spectra was fitted 1:1 in magnitude to the scattering spectra for gaining better visibility into the wavelength dependent trends exhibited by the optical forces compared to the scattering properties for each dimer case. As we can see in Fig. S8a, the spectra of attractive optical forces between the 50 nm radius nanodiscs exhibits a behaviour with wavelength which is close to the scattering properties of the dimer. When the diameter of the nanodiscs is changed to 100 nm, we observe broadening of the spectrum associated with the optical forces and scattering Fig. S8b. For nanobar dimer with thickness 50 nm, side-length 50 nm and length 75 nm, we see that the attractive optical forces follow the scattering spectra closely, as shown in Fig. S9a. When increasing the length of the nanobar to 125 nm, we see broadening of the spectra associated with both the optical forces and scattering Fig. S9b. The spectra of optical forces red-shifts w.r.t the scattering spectra on increasing the length of nanobars, which is also observed in case of nanodiscs on increasing their radius.

9. POLARISATION DENSITY MAP OF NANOCUBE DIMER

The correlation of polarisation density with electric field enhancement was studied in nanocube dimer with 125 nm edge-length and 10 nm inter-particle spacing. Incident light was injected along z-axis with electric-field vector aligned along the inter-particle axis of the cube dimer, i.e x-axis. Fig. S10a shows the E-field enhancement in the cube dimer at resonant excitation conditions 755 nm. The calculation of the polarisation density corresponding to resonant excitation for the dimer at 755 nm shows a magnitude of $14 \times 10^{-9}$ C/m$^2$ as shown in Fig. S10b. The polarisation density drops in magnitude to $10.3 \times 10^{-9}$ C/m$^2$ at 779 nm excitation wavelength S10d. The change in polarisability causes an asymmetry in local-field enhancement along the gap region of the dimer. As it can be seen in Fig. S10c, the hot-spot for field enhancement becomes more prominent around the upper corner of the nanocube dimer. When the excitation wavelength is red-shifted to 809 nm, corresponding to the second peak in the scattering spectra of the nanodimer, the hot-spots of electric-field enhancement shift towards the outer edges of the nanocube dimer, as shown in Fig. S10e. The associated polarisation density for the 809 nm excitation reveals a drop in its magnitude to $4.5 \times 10^{-9}$ C/m$^2$, shown in Fig. S10f. These observations convincingly show the possibility to tune the electric-field enhancement in nanocube dimers by changing the magnitude of polarisation density exhibited by them, which can be controlled by wavelength of the excitation source.
Fig. S7. X-Z view of electric-field profile in gold nanocube dimers under plane wave excitation normal to the plane of nanocubes. Electric-field enhancement in 100 nm edge-length nanocube dimer with 10 nm gap distance at (a) 604 nm excitation, (c) at 670 nm excitation, (e) at 800 nm excitation. Electric-field enhancement in 150 nm edge-length nanocube dimer with 10 nm gap distance (b) at 880 nm excitation, (d) at 862 nm excitation, (f) at 936 nm excitation. The orientation of electric field is along the inter-particle axis of the dimer in x-direction.
Fig. S8. Spectral response of optical forces in nanodisc dimers. 

(a) Response of optical forces with wavelength in nanodisc dimers with 50 nm radius and 10 nm gap distance. 
(b) Response of optical forces with wavelength in nanodisc dimers with 100 nm radius and 10 nm gap distance.

Fig. S9. Spectral response of optical forces in nanobar dimers. 

(a) Response of optical forces with wavelength in nanobar dimers with 50 nm side-length and 10 nm gap distance. 
(b) Response of optical forces with wavelength in nanobar dimer with 125 nm side-length and 10 nm gap distance.
Fig. S10. Correlation of electric field enhancement with polarisation density in nanocube dimers under plane wave excitation normal to the plane of the nanocubes. X-Z view of electric-field enhancement in 125 nm edge-length nanocube dimer with 10 nm gap at (a) 755 nm excitation, (c) 779 nm excitation, (e) 809 nm excitation. Polarisation density for 125 nm edge-length nanocube dimer with 10 nm gap at (b) 755 nm excitation, (d) 779 nm excitation, (f) 809 nm excitation. The electric field was aligned along the inter-particle axis of the dimer in x-direction.
Fig. S11. X-Z view of electric field enhancement and polarisation density in nanobar dimer under plane wave excitation normal to the plane of nanobars. a Local field enhancement and b polarisation density of nanobar dimer at 795 nm excitation. The nanobar dimer had thickness and side length 50 nm and 150 nm length with 10 nm gap.

Fig. S12. Evolution of optical forces with polarisation density strength in nanodimer systems. All values have been calculated at their plasmon resonance excitation conditions respectively. The y-axis in the figure is plotted on the log scale.

10. ELECTRIC FIELD ENHANCEMENT AND POLARISATION DENSITY MAP OF NANO-BAR DIMER

Electric field enhancement and polarisation density were studied under plane wave excitation in nanobar dimer of side length and thickness of 50 nm and a longitudinal length of 150 nm with 10 nm inter-particle separation distance. Light was perpendicularly incident on the plane of the nanobars, and the electric field vector was aligned along the inter-particle axis of the dimer. For resonant excitation condition at 795 nm, we observe a strong enhancement of electric-field in the gap region between the nanobars, as shown in Fig. S11a. The corresponding polarisation density for the dimer at 795 nm excitation reveals a magnitude of $26 \times 10^{-12}$ C/m^2. The polarisation map shows localisation of opposite charges at the facing sides of the dimer, as shown in Fig. S11b, with a symmetric distribution of charge across the nanobar dimer’s surface (along the y-axis).

11. RELATION OF OPTICAL FORCES WITH DIPOLE STRENGTH IN NANODIMERS

The evolution of optical forces with polarisation density is shown for nanodisc, nanocube and nanobar dimer at their respective plasmon resonance excitation condition in Fig. S12. It can be deduced from the data that higher dipolar strengths lead to greater internal optical forces, however there are other important factors like nanoparticle geometry, inter-particle distance,
Fig. S13. Electric field plots of non-paraxial Gaussian beam used as excitation source. a Electric field distribution for a propagating 3D gaussian beam. b Intensity distribution for the Gaussian beam in the x-y plane.

sharpness of corners/edges and symmetry of the nanostructure, which influences the distribution of electrons over the nanoparticle surface, and thus directly influences the optical forces. Moreover, polarisation density also governs the repulsive components of the optical forces, which are not relevant for our case and hence not discussed.

12. SIMULATION METHODOLOGY FOR GENERATING GAUSSIAN BEAM SOURCE

The Gaussian beam source was formulated using COMSOL Multiphysics (Version 5.6) using the non-paraxial approximation [2]. The formulation works on the idea of constructing a beam by addition of waves travelling in multiple directions with the same wave number k. The resulting wave allows the possibility for realising a beam with a waist size which is comparable to its wavelength i.e 1:1. This is however not achievable using the conventional paraxial approximation, since reducing the waist size of the beam results in angular deviation in the beam propagation direction. Fig. S13a shows the side view of a Gaussian beam propagating along the z-axis. The nanoparticle dimer was placed in the focus of the beam at the central position. The alternatively varying electric and magnetic fields are identified from their respective colors on the intensity scale. Fig. S13b shows the intensity distribution of the Gaussian beam in the plane of the nanoparticles i.e. x-y plane. It could be seen that the electro-magnetic field intensity of the beam shows a Gaussian distribution, with maximum intensity localised at the center of the beam which falls exponentially to the outside from all directions.

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