What is it optical binding and how to study this phenomena?

Vítězslav Karásek, Pavel Zemánek
Institute of Scientific Instruments, Academy of Sciences of the Czech Republic, Královo polská 147, 612 64 Brno, Czech Republic

ABSTRACT

Optical binding uses the fact that each object placed into the optical field modifies this field and at the same time interacts with other objects in the field. Under certain circumstances these objects can self-organize and create so called “optically bound matter”. This happens for various sizes of particles and therefore there is no unique method for the theoretical description of the phenomena. If the objects are small compared with wavelength then they can be approximated as single dipoles. On the other hand paraxial wave theory is acceptable for particles with sizes in multiples of the wavelength. We develop a method based on coupled dipole method (CDM) or also known as a discrete dipole approximation (DDA). This numerical method covers the range of sizes when particles are comparable or smaller than wavelength. For some cases the comparison with paraxial wave theory is also possible. Our aim is to specify conditions suitable for creation of optically bound systems which are experimentally verifiable.

Keywords: optical force, optical binding, Bessel beams

1. INTRODUCTION

The phenomenon of optical binding presents results of interactions of light with matter and matter with light. In the example of colloidal particles the particles modify by scattering the incident field which leads to self-organisation of particles to their new positions. Our aim is to find stable positions in which the total forces acting on particles given by incident and scattered fields and the field inside the particles form a stable static configuration in space.

If the forces forming stable configuration are dominant over thermal forces (leading to Brownian motion) then the optical binding can be observed and confirmed experimentally. In order to eliminate all effects coming from mutual interference of incident fields and their non-uniformity we prefer non-interfering Bessel beams. The comparison with Gaussian beams is also presented in this paper.

Although the optical binding was recognised more than ten years ago it remains attractive topic. The reason is that this effect comes from a very sensitive interplay of multiple scattering on all particles involved. This evolves problems in design of experimental setup and uncertainties in detection of particle positions. But bigger problem present theoretical or numerical models of this phenomena. Usually the size of particles is comparable to the wavelength of light and even scattering on a single particle requires strong mathematical background like Mie theory. Unfortunately this analytical method cannot be used for the case of interaction of several particles if self-interaction between particles is to be taken into account. For these purposes we use Coupled dipole method (CDM) also know as discrete dipole approximation. We extended their method for computation with several particles and enabled also computation of forces.

A very good insight into mutual influence of particles offers analytical method. The authors studied the effect of distance between two Rayleigh particles and also calculated the forces acting on them. The particles were located across the incident beam and therefore the incident fields had same phase for both of them. In our setup the particles are located on axis of two counter-propagating beams and differences of phases of the beams between the two particles bring new effects. As the analytical model allows only treatment with Rayleigh particles we can use it only for comparison with our CDM code in the case of small particles although some effects are similar for all sizes.

Further author information: (Send correspondence to P.Z.)

P.Z.: E-mail: zemanek@isibrno.cz, Telephone: +420 541 514 202, Fax: +420 541 514 402
2. THEORY

For the computation of scattering by two particles in general case we use method based on CDM. Although this setup of two beads is the simplest possible arrangement for binding the results are very interesting and show how the binding is very complicated process. Our implementation\(^8\) of CDM allows us to consider any number of particles of any shape and optical properties. It is possible because the numerical method divides all particles into sufficiently small parts which may be approximated as single dipoles. The mutual interactions of all dipoles result in approximated distribution of dipole polarisation in all the particles. From the distribution of polarisation we calculate forces acting on individual particles.\(^9\)

2.1. Coupled dipole method

Let us consider for simplicity only one scatterer which is divided by cubic lattice into \(M_x M_y M_z\) dipoles. These natural numbers reflect the size of the scatterer, i.e. the maximal extend in \(x, y,\) and \(z\) direction is smaller than \(M_x d, M_y d\) and \(M_z d\), where \(d\) is the lattice constant. The upper limit for separation \(d\) of neighbouring dipoles is:

\[
d \leq \frac{\lambda}{20|m|},
\]

where \(\lambda\) is the wavelength of light in the medium and \(m\) is relative index of refraction between the scatterer and the surrounding medium. This limit is tested by comparison of results with several number of dipoles. The dipoles are disturbed by the incident fields and are sources of the secondary radiation. By this radiation they influence each other and the result of this influence is the distribution of total electric field in the scatterer. The method has several versions differing mainly in the manner of division of the scatterer into dipoles and a number of such dipoles. We adapt a method described in [7,11,12] and in [9] for calculation of forces acting on particles. This method uses convolution and FFT techniques to find a solution of the interaction equation (4) which describes mutual influences of incident field and dipoles themselves. The division used in this method requires a strict location of dipoles which must possess a cubic lattice with the lattice constant \(d\). Each dipole is described by a vector-index \(i = (i_x, i_y, i_z)\) and therefore its position is \(r_i = id + r_0\), where \(r_0\) is the location of the first dipole. The geometry of the scatterer is described by \(i_x = 0, 1, \cdots M_x - 1, i_y = 0, 1, \cdots M_y - 1\) and \(i_z = 0, 1, \cdots M_z - 1\). The goal of CDM is to find polarisations \(p_i\) which are connected with total (macroscopic) field \(E_i\) by polarisability \(\alpha_i\):

\[
p_i = \alpha_i E_i.
\]

The total field \(E_i\) consists of the incident field \(E_{\text{inc},i}\) and fields scattered from all other dipoles. The scattered field at \(r_i\) from dipole \(j\) with polarisation \(p_j\) is given by \(-A_{ij} p_j\), where the radiation matrix \(A_{ij}\) has a form:\(^{13}\)

\[
A_{ij} = \frac{\exp(i k r_{ij})}{4\pi \varepsilon_0 \varepsilon_r r_{ij}}
\times \left[ k^2 (n_{ij} n_{ij} - 1) + \frac{ik r_{ij} - 1}{r_{ij}^2} (3 n_{ij} n_{ij} - 1) \right],
\]

where \(k\) is a wavenumber of the light in the medium, \(r_{ij} = |r_i - r_j|\), \(n_{ij} = (r_i - r_j)/r_{ij}\) and \(1_3\) is \(3 \times 3\) identity matrix. Therefore the total electric field \(E_i\) at \(r_i\) is expressed as

\[
E_i = E_{\text{inc},i} - \sum_{j \neq i} A_{ij} p_j,
\]

substitution from (2) for \(E_i\) gives us the set of linear equations for desired \(p_i\). As it was outlined before this set must be solved iteratively by conjugate gradient or bi-conjugate gradient methods.\(^{14}\) These methods require calculations of the matrix-vector products \(Ap^{(k)}\), where \(p^{(k)}\) is a \(k\)-th guess of polarisations. The number of numerical operations needed for such a product is proportional to \(N^2\), where \(N\) is the total number of dipoles usually in range from thousands to millions. Another problem brings memory storage requirement of matrix
which are equal to $144 \times N^2$ bytes. However from the requirement of location of dipoles on cubic lattice
the difference between dipoles $i$ and $j$ is $r_{ij} = d(i - j)$ and therefore the $3 \times 3$ matrices $A_{ij}$ depend only
on the difference $i - j$. Primary this fact significantly reduces memory requirements and secondary enables
computation of matrix-vector products $Ap^{(k)}$ as convolutions which are computed according to convolution
theorem\textsuperscript{14} by FFT algorithm.\textsuperscript{12} The number of numerical operations is in this case proportional to $N \log N$
only.

2.2. CDM applied to several objects

In application of FFT in CDM we must compute with all dipoles forming the cuboid with shape $M_x \times M_y \times M_z$.
Therefore a scatterer of arbitrary shape must be enveloped by $M_x \times M_y \times M_z$ cuboid in order to be described.
Since we analyse a system of two or more particles with various separations, using one cuboid for this system
would be very ineffective because of a very large number of unoccupied dipoles. Therefore we tried to extend
the method also for computation with several objects.\textsuperscript{8} The principle is the dividing the total field in the first
sphere into field from the dipoles belonging to this sphere (local field) and field form dipoles of the other sphere
(distant field).

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig1.png}
\caption{An example of modelling two spheres with their lattices separated by $Q$.}
\end{figure}

\begin{equation}
\mathbf{E} = \mathbf{E}_{\text{loc}} + \mathbf{E}_{\text{dist}}.
\end{equation}

We have shown\textsuperscript{8} that the distant field can be also computed using FFT because for mutual distances between
dipoles belonging to different spheres $r_{ij}$ depends only on the difference $i - j$:

\begin{align*}
   r_{ij} &= r_i - r_j = d(i - j) + Q, \\
   r_{ij} &= \sqrt{d^2 |i - j|^2 + |Q|^2}, \\
   n_{ij} &= r_{ij}/r_{ij},
\end{align*}

where $Q$ is the displacement vector between two spheres as is demonstrated in Fig. 1.

2.3. Calculation of optical forces using CDM for several objects

There exist two CDM methods suitable for computing of forces between dipoles. The first developed by
Draine and Weingartner\textsuperscript{7} computes the force from time-averaged rate of transfer of momentum by the scattered
radiation. The second method was developed by Hoekstra et al\textsuperscript{9} and uses differentiation of field for computing
the force. They showed the equivalence of these two methods. We use the second one because it is more suitable
for bigger number of dipoles. We modified it for the case of several objects using the same principle as in the case of computing field. Time averaged force acting on a dipole $i$ can be written as:

$$\langle F_i \rangle = \frac{1}{2} \Re \left[ (p_i \cdot \nabla)E_i + i\omega p_i^\ast \times B_i \right]. \quad (5)$$

The principal step is the splitting of the total force acting on dipoles into forces acting from the incident field and the scattered field.\footnote{2} If the incident field can be written in an analytical form (such as plane wave, Gaussian beam, Bessel beam), we can easily differentiate it and evaluate by (5).

The differentiation of the scattered field is done by differentiation of radiation matrix $A_{ij}$. For the number of dipoles used this numerical operation must be made again by FFT.\footnote{9} Transitional symmetry in expressions for forces between dipoles of different objects is preserved, too. So we extended computation of forces for several objects in the same manner as for computation of field.

3. BINDING IN TWO COUNTER-PROPAGATING BEAMS

![Figure 2. The demonstration of Bessel beam generation.](image)

We use our model to study the binding effects of two dielectric spherical particles (beads) in two counter-propagating, non-interfering Gaussian or Bessel beams. For the purposes of studying binding (as will be explained later) is more appropriate to work with Bessel beams. These beams feature a central core of high intensity and so they are suitable for 2D confinement of particles. Moreover their homogeneity in the direction of propagation provides simplified on-axis behaviour of particles, because the particles in this direction do not suffer from gradient forces which are present in Gaussian beams. Therefore if a number of particles is placed into such axially independent field, all the axial forces are purely the binding forces. We mainly studied how the force interaction coming from the light scattering between the beads depends on their size and mutual distance. Moreover we studied the influence of the core size of the Bessel beams which plays very important role in the particle behaviour. The Bessel beams in our setup are created by an interference of plane waves emerging from the axicon (see Fig. 2). All the plane waves behind the axicon have their wave vector tilted to the optical axis by angle $\alpha_0$. By interference of all these waves there is created the Bessel beam of zeroth order. The derivation of the approximative relation can be found in.\footnote{15} For small angles $\alpha_0$ their intensity profile in the plain perpendicular to the optical axis is mainly given by Bessel function of zeroth order

$$J_0^2(k \sin(\alpha_0) \sqrt{x^2 + y^2}). \quad (6)$$
The roots of Bessel function $J_0$ determine the places of zero field. Their radial distance $\rho_0$ from the axis is given by:

$$\rho_0 = \frac{R_0}{k \sin \alpha_0}, \quad (7)$$

where $R_0$ are the roots of the Bessel function of zeroth order

$$J_0(R_0) = 0, R_0 \approx 2.4048, 5.5201, 8.6537, \ldots$$

Therefore the quantity $\rho_0$ is called the radius of the core of the Bessel beam and is connected to the angle of the Bessel beam $\alpha_0$ by relation (7).

### 3.1. The modeling of binding of two beads

In our numerical model we assume the simplest possible case of two beads in two counter-propagating non-interfering Bessel beams. The beads are located on the common optical axis of the incident beams. For various distances we compute the forces acting on the beads. We made the simulations for various sizes of the beads and radii of Bessel cores thus studying the effect of these parameters on binding. The illustration of the setup is in Fig. 3 together with computed results.

![Diagram of two beads in Bessel beams](attachment:bead_setup.png)

**Figure 3.** Schematic view of numerical simulation of binding with computed forces and separation works. The results are for dielectric polystyrene beads ($n = 1.59$) of radii $a = 250$ nm in water. The radius of the Bessel beam core $\rho_0$ was $1.6 \mu$m, wavelength ($\lambda = 800$ nm in water) and intensity of the beam on the axis was by way of illustration chosen to intensity of Gaussian beam with power $1$ W focused into waist of radius $1 \mu$m. The location of beads in which the resulting forces are zero and separation work is minimal corresponds to stable position of beads resulting from optical binding.
3.2. The classification of forces
As is displayed in Fig. 3 we assume two beads labeled as A and B which are under influence of two non-interfering Bessel beams titled as Left and Right. The forces from incident fields are separated into four parts: Left beam acting on the A bead, right beam acting on the A bead, left beam acting on the B bead and right beam acting on the B bead. The forces from incident fields are also separated into four parts: A bead acting on itself, A bead acting on B bead, B bead acting on itself, B bead acting on A bead. As the configuration is completely symmetrical the total forces acting on the beads A and B are opposite and we can only analyse forces acting on bead B.

3.3. The comparison of Bessel beams with Gaussian beams
For the case of binding is important the dependence of the constituent forces on the separations of the beads. The detailed view on this dependence is in Fig. 6 with comparison between Gaussian and Bessel beams. The waists of counter-propagating non-interfering Gaussian beams are both right in the middle between beads. The size of waists and radius of cores were chosen in order to the beams having similar properties(intensity and power in the core). In the graphs we can see, that the positive sign for the force from the left beam and negative sign for the force from the right beam according to their directions. But the dependence of the force from the right beam is up to some small fluctuations very flat for the Bessel beams in contrary to the Gaussian beams. Although the order of this difference is small comparing to the amplitude of the forces this deviations and fluctuations play very important part in the formation of total forces. The total effect of the forces from incident beams is pushing the beads apart. For distant beads the amplitudes of these force tend to be the same according to the expectations. Their sum for the case of Bessel beams goes to zero.

The forces by which beads act on each other are smaller and the beads attract each other. So the A bead attracts B bead moreover the B bead pulls itself toward A bead. For distant beads amplitudes of these forces tend to zero. The sum of both forces is in magnitude very similar to the forces from incident beams up to small difference which shows up in the shape of the total force. From these follows that the total forces are very sensitive result of interplay of forces coming from incident and scattered fields. This is very interesting result because the forces from incident fields come from the “first order of scattering” while the forces from scattered field reflect the interaction of scattered field with itself thus being of the “second order”.

3.4. The binding in the Bessel beams
As was demonstrated in Fig. 6 the axial non-homogeneity in Gaussian beams has very big effect on the creation of stable positions. Moreover we cannot in principle distinguish the effect of multiple scattering between beads from the effect of non-uniformity of the field. The Bessel beams are even in practical realisations by axicon very uniform along the optical axis. In the next part we will therefore focus on this type of beams.

The core size of the Bessel beams plays crucial role in the creation of the stable positions. For smaller radii of the Bessel beams there exist more regions of beads’ separations. As seen in Fig. 5 this effect is very sensitive on the core size. For a given core of the Bessel beam the beads of all sizes have roughly similar behavior of the forces on their separations. For the bigger sizes of beads the beads are closer in the binding positions. Moreover the size of beads may alter the existence of the binding positions for certain separations. This effect is still partially unexplained and is related to distribution of polarization inside the beads.

From a closer look on the dependence (Fig. 4) of the force on the separations of the beads is apparent that the shape of the force is given by wavelets. The reason for these wavelets is following: The incident fields along the optical axis propagate with different k-vector $k_z$ than field radiated by the beads. The radius of the core $\rho_0$ related to angle of the plane waves in the Bessel beam $\alpha_0$ by (7) does not only determine the radial extent of the Bessel beam but also changes the value of axial part of wave vector

$$k_z = \frac{1}{\rho_0} \sqrt{k^2 \rho_0^2 - R_0^2}. \tag{8}$$

We now deduce the wavelength of these wavelets. Let us now for the simplicity of explanation forget the left beam. This reduction does not influence the following explanation because right and left do not interfere. The
Figure 4. The existence of force wavelets is presented with Bessel beams ($\lambda = 800\, \text{nm}$ in water, $\rho_0 = 1.6\, \mu\text{m}$). There is represented an area where beads of all sizes have stable positions. The extent of this area for individual sizes of beads depends very strongly on the “wavy character” of the dependence of the total force on the distance of spheres. The waviness shows very sensitive dependence on the beads size. For beads with size of radius $a = 250\, \text{nm}$ vanishes but for slightly bigger or smaller spheres is present. The magnitude of waviness increases with radius to maximum value at $a = 350\, \text{nm}$ and then decreases towards second minimum for beads of radius $a = 420\, \text{nm}$ where the waviness again disappears.
change of phase of the right beam with position is given by value of $k_z$ given by (8). The polarizations of dipoles of $B$ bead are given by incident right beam and by dipoles belonging to $A$ and $B$ bead. But the field from $A$ to $B$ radiates by $k$ which is different from $k_z$. The forces on individual dipoles are given by multiplication of polarizations of dipoles with derivatives of field as follows from (5). In the multiplication are also terms proportional to:

$$\exp^{ik_z} \exp^{i(k+k_z)z}, \tag{9}$$

and from this follows that wavelength of the wavelets is given by:

$$\lambda_{\text{wav}} = \frac{2\pi}{k + k_z} = \frac{\lambda \lambda_z}{\lambda + \lambda_z}, \text{ where } \lambda_z = \frac{2\pi}{k_z}. \tag{10}$$

Simple tests for different beam cores confirmed agreement of this formula with calculated results. Therefore the force wavelets have origin in the interference of incident beams with radiated fields. The reason for the dependence of the waviness on the beads’ size is for us still bit unknown, but it clearly reflects the distribution of the polarizations inside the beads.

4. CONCLUSION

The analytical studies of optical binding are only possible for particles of Rayleigh size. But the binding forces are very weak in comparison with thermal motion for this size of particles. We applied analytical method presented by F. Depasse to our setup and got very good agreement with method based on CDM. Our method is mainly useful for particle sizes comparable with wavelength. Other methods used for example in [5] are applicable only for particles of bigger sizes. We made calculations for this case too and got partially good agreement with the experiment. Anyway the numerical results showed that binding is very sensitive process and that even small uncertainty in parameters of setup or particles may give deviation in the result.

In this article we presented that Bessel beams are very appropriate for the study and experiments with optical binding. Their main advantages are uniformity of the field along optical axis and radial localisation of the field in the core. The simulations showed that size of the core is very sensitive parameter for resulting separations of optically bounded beads. This fact may complicate the experiments which we plan to do in the near future. Moreover from the results follows that binding has very complex dependence on beads size. This is induced by the distribution of the polarizations inside the beads which is the next target of our research.
Figure 5. The dependence of the total forces on the radii of the beads and the Bessel beam cores. The separations of the beads in the stable positions are marked by bigger dot. Two beads separated by this distance do not feel any optical force and when they are deflected from this stable position the emerged optical forces put them back. The simulation is made with polystyrene beads ($n = 1.59$) placed in water where the wavelength of the light is 400 nm.
Figure 6. Comparison of dependence constituent forces between two counter-propagating Bessel beams and two counter-propagating Gaussian beams on the distance between the beads. The wavelength of the beams was chosen to 800 nm in water. The intensities of the beams on the axis were chosen to be the same. The results are for dielectric polystyrene beads \((n = 1.59)\) of radii \(a = 250\) nm in water. Although the differences in constituent forces between Bessel and Gaussian beams are very slight, the total forces which are sums of these forces are different. For the case of Bessel beams it leads to stable positions with beads separation 12 \(\mu\)m.
ACKNOWLEDGMENTS

This work was partially supported by the European Commission 6'th framework programme – NEST ADVEN-
TURE Activity – through the Project ATOM-3D (508952), and by Ministry of Education, Youth and Sports
of the Czech Republic (project No. LC06007).

REFERENCES

1236, 1989.
3. S. A. Tatarkova, A. E. Carruthers, and K. Dholakia, “One-dimensional optically bound arrays of micro-
5. N. K. Metzger, K. Dholakia, and E. M. Wright, “Observation of bistability and hysteresis in optical binding
7. B. Draine and J. C. Weingartner, “Radiative torques on interstellar grains i. superthermal spin-up,” Astro-
11. B. Draine, “The discrete-dipole approximation and its application to interstellar graphite grains,” Astro-