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Jonathan M. Taylor

Optical Binding Phenomena: Observations and Mechanisms

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Supervisor's Foreword

Optical binding is a remarkable phenomenon whereby the presence of a micro-particle in an optical landscape can influence the light distribution which in turn can affect the position of the particles. As a simple example two laser beams pointing towards each other a counter propagating trap form a potential well analogous to springs attached to particles. If one increases the number of particles in the trap then naively one would expect all the particles to collect in the centre of the well. However, the effect of optical binding means that the presence of one particle affects the distribution of light with the effect that the particles can arrange themselves into arrays as well as displaying other interesting phenomena. Optical binding is both of theoretical interest and has applications in micromanipulation and assembly.

The work described in this Thesis combines experimental results with a sophisticated generalized Lorenz–Mie scattering model implementation developed by Jonathan Taylor as part of his Ph.D., the capabilities and speed of which make it possible to study time evolution of a multiple particle system. The work begins with an introduction to generalized Lorentz–Mie scattering theory written in a manner accessible to the non-specialist, and including some novel results. It then compares experimental and theoretical results for a range of trap configurations, not just demonstrating agreement between theory and experiment, but using the model to provide insight and interpretation of the experimental results.

This thesis therefore contains results of the full spectrum of physics activities: rigorous electromagnetic theory, advanced computer simulations, extremely sensitive experimental results, and physical analysis and interpretation of those results. For example, he has shown that particles can undergo circulatory motion in a trap where all the parameters are constant. This remarkable result was described both by his computer model, and by a simple heuristic model showing an intuitive explanation, as well as being observed experimentally.

Durham, March 2011 — Dr. Gordon D. Love

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Most of all I would like to thank my supervisor Gordon Love for his support and guidance throughout the four years work leading to this thesis, for having confidence in the unexpected tangents the project took...and indeed for persuading me to work on optical trapping in the first place, rather than on his "bait" Ph.D. project!

I am also very grateful to the various people I have been fortunate enough to collaborate with or correspond with, including Colin Bain, Pavel Zemánek, Jack Ng and Reuven Gordon.

Finally, I must acknowledge the "help" of the various inhabitants of room 125b, a room which has seen some great mysteries as well as great engineering achievements, and the invaluable assistance of MH who had a beer ready the one time when I most needed it.

Contents

Chapter 1
Introduction

1.1 Motivation

This thesis investigates phenomena occurring when multiple particles are confined in the same optical trap, leading to light-mediated interactions between the trapped particles (*optical binding*). These interactions are not only of interest in terms of the fundamental optical physics involved, but also have many practical implications for micro-manipulation of dielectric particles. Multiple particles may be manipulated for the purpose of microstructure construction, using either:

- individual optical tweezers, where any inter-particle interactions are an undesirable side-effect [1]; or
- broader optical fields, where inter-particle interactions can be critical to the self-assembly of a structure [2–4]

Multiple particle interactions are also of interest in optical lattices, optical sorting and optical transport [5–8].

The aim of this thesis is to develop a better understanding of the physical mechanisms underlying the optical binding interaction. The intention, where possible, is to discuss the phenomena in a physically intuitive manner, drawing insight from a rigourous analysis of the system to develop simple, easily-understood explanations for the effects observed.

This thesis presents results on the optical binding of optically-trapped microparticles. A sophisticated Mie scattering model is developed, capable of performing time-evolution simulations of a multi-particle system. This is used to analyse and interpret experimental results in evanescent and Gaussian beam traps, and to develop simple, intuitive explanations for the observed phenomena. Novel trapped states are reported, that do not conform to the symmetry of the underlying trap. A common theme throughout this thesis is the "emergent" phenomena that occur when multiple particles are trapped together, which cannot easily be predicted by considering each particle in isolation.

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1.1.1 Experimental History of Optical Trapping and Binding

Optical confinement of micron-sized particles in two dimensions using a focused laser beam was first demonstrated by Ashkin et al. in 1970 [9], and subsequently extended to three-dimensional confinement using two counter-propagating beams, originally in the form of an ion trap [10]. A significant development from this has been the field of *optical tweezers*, where a high numerical aperture laser beam is used to trap and manipulate a microparticle. A relatively independent development has been the investigation of interactions between multiple trapped particles (*optical binding*, generally in low numerical aperture configurations).

The term *optical binding* was first introduced in [11], referring to interference effects between the light scattered by a single trapped particle and the background laser trapping light. This interference strongly modifies the electromagnetic field around the particle, and the field experienced by a second nearby particle (and hence the force on it) is different to that which either of the individual particles would experience in isolation. The presence of interference fringes produced by the scattered light tends to cause particles to be trapped and "bound" at discrete inter-particle distances that, depending on the trap geometry, will often be multiples of the laser wavelength (lateral geometries) or half the laser wavelength (longitudinal geometries).

"Optical binding" has more generally been used to refer to any experimental phenomenon whereby multiple trapped particles interact to form well-defined, reproducible, bound structures. We will see in Chap. 4, however, that in some cases the phenomena are not in fact caused by binding in a strict interference-based definition, as was the case in [11].

This thesis is almost exclusively concerned with optical binding effects, rather than optical tweezers, but we will return to this distinction in Chap. 5 where we will discuss the implications of optical binding for high numerical aperture optical tweezers experiments. Consequently the brief review of the field given in the following sections will focus on optical binding, and areas of optical tweezing where inter-particle interactions are expected to be relevant. In addition to this, subsequent chapters give a further introduction to the literature relevant to the chapter topic.

1.1.1.1 Optical Binding

Optical binding between multiple optically trapped particles was reported by Burns et al. in 1989 in a lateral configuration where the particles lay in a plane perpendicular to the direction of propagation of the trapping laser beam [11, 12]. In this case the confinement was only two-dimensional. Interestingly, this lateral configuration has never to our knowledge been extended to three dimensions by

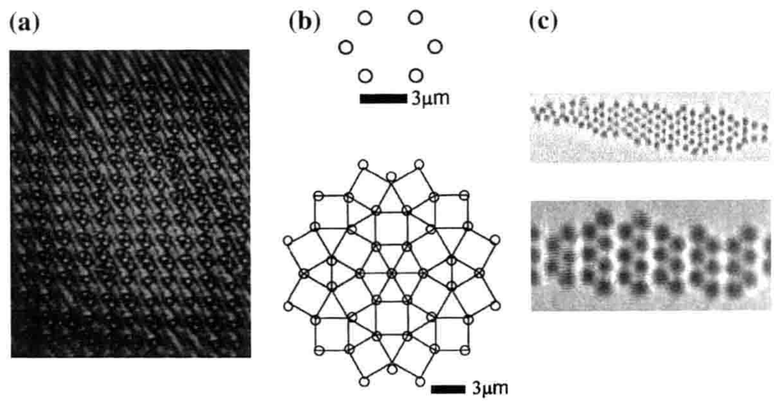

Fig. 1.1 Examples of optically bound particle clusters (**a**), two-dimensional arrays in [12] (**b**), relatively widely-spaced "molecules" in [13] (**c**), fairly close-packed regular structures in an evanescent wave trap, in [20]

the use of two counter-propagating beams[1], although such a configuration has been studied theoretically [13].

It was not until 2002 that two groups [14, 15] independently reported longitudinal interactions between multiple particles in a counter-propagating Gaussian beam trap (where the particles lie parallel to the direction of propagation of the trapping beams). This is the type of configuration that we will mostly focus on in this thesis.

Optical binding has been observed in theory and experiment to give rise to a rich tapestry of nonlinear static and dynamic behaviour, including:

- chain formation [14–16], where trapped particles in a counter-propagating Gaussian beam trap form linear chains, with the spacing between the particles dictated by the optical binding interaction.
- bistability [17, 18], where for some experimental parameters there are multiple stable spacings between a given number of particles. For a Gaussian beam trap this generally means one "standard" spacing, which can be explained using our model in Chap. 4, and one very close spacing where the particles are almost touching, where the repulsion is produced by complicated near-field effects.
- two-dimensional "crystal" arrays [12, 13, 19–21], where sub-micron-sized particles form two-dimensional regular structures perpendicular to the direction

[1] This may be because there are many interference fringes formed, with each of these defining a different plane of trapping perpendicular to the beam axes. In order to form optically bound clusters in a single plane there must be some way of ensuring all the particles are located in the same plane. This would probably require manual loading using "helper" optical tweezers.

of propagation of the trapping beam(s). These have been observed in a number of different situations (see Fig. 1.1):

- broad Gaussian beam [12] (pseudo-plane wave): fairly close-packed regular structures trapped in two-dimensions, with the third dimension of confinement provided by the walls of the cell;
- counter-propagating plane waves[13]: theoretical prediction of two-dimensional "molecules" formed from particles which are several diameters apart;
- evanescent wave trap [19–21]: fairly close-packed regular structures trapped in two dimensions in an evanescent wave, with the third dimension of confinement provided by the totally internally reflecting substrate.

As we will discuss in Chap. 3 these scenarios are particularly challenging to understand, not only because of the large numbers of interacting particles but also because of the combination of optical binding, physical close-packing constraints and possible electrostatic charges, all of which can have a significant influence on the structures formed.

- periodic particle motion and instabilities [13, 22–25] where, despite the overdamped nature of the system, driven harmonic oscillations and instabilities can be observed.

Possible future areas of development for optical binding include:

- aerosol trapping: recent preliminary results have reported optical trapping and binding of aerosol particles in air [26–30].
- large space-based structures: it has been proposed that the self-organising properties of optically bound particles could be use to self-assemble enormous planar structures in space, which could be used as telescope mirrors or solar sails [3]. Although this would be a very exciting and spectacular application for optical tweezing and binding, our findings presented here cast doubt on the scalability of this proposed technique. We will return to this question in Chap. 5.

1.1.1.2 Optical Tweezers

Confinement of a particle using a single-beam high numerical aperture optical tweezers set up was demonstrated in 1986 [31]. Since then optical tweezing and micromanipulation has developed into a broad field with many applications. Comprehensive reviews of micromanipulation can be found in [7, 32].

In the context of the phenomena discussed in this thesis, our main interest in optical tweezing is where multiple particles are being trapped together. Examples include:

- Time-sharing to produce multiple optical tweezers from a single laser beam [33]. This is a simple system to design, but suffers from scalability problems, since as more trapping sites are added the fraction of time that the laser

is assigned to each trapping site is reduced, and the number of trapped particles is limited by the field of view of the trapping objective lens. This reduces the strength of the trap and increases the tendency for particles to diffuse out of the trap. It does however have the advantage that, since only one particle is illuminated at a time, optical binding effects should be negligible.
- Holographic optical tweezers which can generate arbitrary "optical landscapes" [5, 6]. Particles will interact with the background optical field and can be trapped. If the particles are close enough together, optical binding interactions may be extremely important in such a setup. An example of a simple optical landscape is the interference fringes discussed in Chap. 3, for which we show that optical binding effects are absolutely critical to understanding the trapped structures which form.
- Measurement of hydrodynamic interactions between multiple trapped particles [34, 35]. This is of dual interest in the context of this thesis: an accurate description of hydrodynamic interactions may be important for a detailed understanding of close-packed trapped structures (see Chap. 3), and when performing hydrodynamic measurements it is important to understand whether optical binding effects may be present because, if so, this needs to be taken into account in the analysis of the experiment.
- Bessel beam guiding. The non-diffracting nature of Bessel beams make them useful for trapping and transport of multiple particles [36], and also enables easy trapping and transport of multiple particles [37, 38].
- Extended trapping regions such as those generated by vortex-like traps [39], where there has been little attention paid to any optical binding interactions that may be present between multiple particles.

In many such optical tweezers arrangements, optical binding is not an intended consequence of the setup. Rather, it is seen as an inconvenience—if indeed it is considered at all. As the numbers of trapped particles (particularly particles with higher radii and refractive indices) increases, the potential for significant optical binding effects will increase.

1.1.2 Interpreting Binding Experiments

Although there have been intermittent attempts to theoretically model the optical binding behaviour between multiple trapped particles, this has proved extremely challenging. One of the main challenges is knowing the experimental parameters well enough to carry out a simulation which can reasonably be expected to agree with experiments.

McGloin et al. developed a simple model based on consideration of the scattering force produced by the light "refocused" by successive particles in a chain [18, 40, 41]. Although we have revisited and extended such a model in Chap. 4, there do appear to be shortcomings with the comparison of experiment and theory presented in [40]. Reasonable agreement was presented between their experimental results and their theoretical model, but in contrast Mie scattering theory predicts significantly different results for the experimental parameters quoted in that paper (different particle spacings, or even whether stable chains are supported or not). This highlights the sensitivity of optical binding experiments to the exact experimental parameters, some of which are often hard to directly measure.

Direct visualization of the modification to the background laser field was demonstrated in [16], using two-photon fluorescence to directly image the intensity of the electromagnetic field around chains of trapped particles. Here good agreement was shown between a paraxial field propagation model and the experimental results. Unfortunately a lack of detail in some of the parameters used has meant we have been unable to make a direct comparison between their model and our Mie scattering model.

It was only in 2008 that very good agreement was demonstrated between Mie scattering theory and experimental results from fibre-based optical trapping experiments at the University of Victoria, Canada (Gordon et al. [24, 42]). It is interesting to note that this significant milestone was achieved using a fibre-based trap for the experimental measurements: this considerably reduces the challenges of alignment and beam quality. It is these factors which prove a considerable obstacle to accurate quantitative measurements in lens-based counter-propagating beam traps.

In addition to this, a model based on coupled dipole calculations has also recently been used to inform a simple explanation of the mechanisms that led to chain formation in a counter-propagating Bessel beam trap [37]. It is worth noting that this mechanism is very different to that which we discuss for Gaussian beam traps in Chap. 4, despite the apparent similarity in the trap configuration and nature of the particle chains formed.

The results of Gordon et al. have validated the use of Mie scattering theory as the gold standard in the modelling of optical trapping and binding, and that achievement in [42] provides important support for our choice of techniques used in this thesis, as well as providing an independent comparison for our Mie scattering computer code, developed separately to theirs. The motivation behind this thesis is echoed by Gordon et al.:

> *Currently, no theory has explained fully the occurrence of inhomogeneous particle spacing, both for a particle number dependency and a dependence on inter-array particle positions (i.e. inner and outer inter-particle spacings differ for a fixed number particle array), and the spontaneous onset of oscillations observed in the dual beam trap* [24].

Chapter 4 of this thesis will address all these questions.

1.2 Synopsis

The aim of this thesis is to offer physical interpretations and insights into optical binding phenomena observed in optical traps. The common theme throughout is the concept of interactions between multiple particles, and the fact that they give rise to behaviours which are substantially different from the behaviours of isolated trapped particles. The observations discussed here cannot be predicted simply by studying isolated particles.

Some of the experimental results which form the starting point for the investigation are from our own experiments. The experimental results referred to in Chap. 3 are the work of our collaborators (whose contribution is made clear in the text where this is the case). We analyse and interpret these results using Mie scattering theory. We developed the computer model used in order to have code which meets the needs of the analysis. In addition to Mie scattering simulations, the results in Chap. 4 are interpreted using a simpler conceptual model which offers more insights into the physical mechanisms underlying the effects.

Chapter 2 describes Mie scattering theory, and briefly discusses our computer model. Some of the content of this chapter draws together results published by a variety of authors into a coherent whole, and some of the results and approaches within the chapter are entirely novel work. The distinction between these is made in the introduction to the chapter, which also briefly summarizes the history of Mie scattering theory and its generalizations.

Chapter 3 discusses evanescent wave trapping experiments, and the two-dimensional "crystal structures" of nanoparticles reported by Bain et al. [19, 20] which are formed by optical binding effects. We explain some of the structures through Mie scattering theory, and discuss the relative contributions of optically-induced forces and collisional interactions on the nature of the structures which are formed.

Chapter 4 considers a trap formed by two counter-propagating Gaussian beams. We describe new experimental results showing trapping configurations which do not conform to the underlying symmetry of the trap, resulting in both stationary modes and non-stationary trapping modes in which particles circulate around the trap away from the common beam axis. We show that such configurations are predicted by Mie scattering theory, along with the more familiar simpler on-axis stationary trapped chains. We discuss the physical interactions which give rise to each of these configurations, and show that the stationary chains can be fairly well described using an extremely simple conceptual scalar model of the light-mediated interaction.

Finally, Chap. 5 summarizes the conclusions drawn from this work, and topics worthy of further investigation.

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