QUANTUM-OPTICAL TRAPPING OF NANODIAMONDS CONTAINING NV CENTRES

By

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Typeset in $\mathbb{E} T_E X 2_{\mathcal{E}}$.

Except where acknowledged in the customary manner, the material presented in this thesis is, to the best of my knowledge, original and has not been submitted in whole or part for a degree in any university.

Reece P. Roberts

Acknowledgements

Ten months ago I started this project, which turned out to be a large battle against very tiny particles. This thesis is the report of the work that was completed, however it cannot express the trials and tribulations along the way which include long days, aligning, adjusting, building and then bumping the finely tuned optical elements in the lab. Neither does it express the refreshing delight when the experiment falls into place and a little bit more is learned from the system. Unfortunately the joy of learning does not like to hurry. Alas more joy is found from those who have taken this long journey with me.

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Abstract

Optical trapping offers a non-contact, non-destructive tool for manipulating and handling particles from micron sized particles down to individual atoms with light. One can distinguish two different applications to optical trapping. The first relying on the ability to trap small particles (from tens of nm to tens of μ m); and the second one related to the confinement and cooling of atoms or collections of atoms. Until now there has been no system that combines the forces due to both classical trapping and atom trapping as they are usually contained in completely separate parameter regimes even though both of these trapping applications arise from the same force. The force is the result of the interaction of the polarisability of the trapped object, which depends directly on the dipole strength, with the electric field of the trapping laser. In the case of classical trapping, the force acts on an induced dipole caused by the electric field on the object, whereas, in the case of atom trapping the induced dipole arises from the transitions present in the electronic structure.

In this way, optically trapped nanoparticles containing embedded optical defects brings about new regimes of trapping and cooling nanoparticles. This new quantum-optical trap allows well established techniques from atom trapping to be applied to the nanoparticles. The result is a quantum-optical trap showing capabilities with much higher trapping strengths and therefore enhanced control for manipulation. The quantum-optical trap also leads to the direct characterisation of the solid state transition properties of the embedded optical defects through force measurements of the trapped nanoparticle. In addition, in the future by applying well established techniques from atom trapping and cold atom physics to the optically trapped particle we can produce a dramatically stronger trap with the potential to cool the nanoparticle centre of mass motion down to its quantum ground state.

In this thesis I focused on identifying the trapping forces on the optical defects inside of optically trapped nanoparticles. I developed an experimental setup and procedure to isolate, trap and reliably measure the trapping behaviour of nanoparticles as a platform for investigating the quantum forces on optical defects in nanodiamonds, specifically the quantum force due to the 1042nm metastable transition of the nitrogen vacancy centre.

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Introduction

1.1 Background and scope

The concept of optical trapping originated in 1970 when Arthur Ashkin of Bell Laboratories first theorised and experimentally proved that a tightly focused beam of light would result in a radiation pressure that would push and accelerate tiny particles. He observed that not only could the focused beam accelerate the tiny particles but they could also be trapped within the tightly confined focus of the light beam. Arthur Ashkin published a series of seminal papers outlining his observations along with an accompanying classical theory explaining his results [1–3]. These papers not only demonstrated optical trapping in aqueous mediums, but also optical trapping in air (optical levitation). Since these ground-breaking discoveries, optical trapping was taken up in a wide range of applications especially in the biological sciences with the manipulation of living cells [4].

Atom trapping is actually also a result of Ashkin's work in the 1970's. Since classical optical trapping was established it was predicted that similar effects were possible with individual atoms. The main difference between atom trapping and classical trapping is that the force is acting on the dipole of an optical transition instead of acting on an induced dipole cause by the incident electric field. It wasn't until 1987 when Chu et. al. demonstrated the first working optical trap for neutral atoms [5]. Cooling and trapping atoms were both developed concurrently and in 1997 the Nobel Prize was awarded to Steven Chu, Claude Cohen-Tannonudji and William D. Phillips for development of methods to cool and trap atoms with laser light. The methods they developed to produce colder and colder atoms in order to provide more stable atom traps include Doppler cooling [6], optical molasses [7], velocity selective coherent population trapping [8] and Sisyphus cooling [9]. The techniques they developed enabled further discoveries which resulted in the field of cold atom physics. Applications that arose from cold atom physics include; producing one-component plasmas [10], producing Bose-Einstein condensations in a dilute gas [11], measuring quantum

properties on single atoms [12] and increasing precision of atomic clocks [13].

Recently optical levitation is being investigated for its potential applications in the field of quantum information and computing. However, in the early days of optical trapping, optical levitation did not immediately find such a rich field of applications as did trapping in fluid find in the biological sciences. This is chiefly due to the difficulties in producing stable traps. Optical trapping in an aqueous medium has the advantage of the viscosity of the fluid that reduces the motion of the particle to a point where it can be easily trapped at the focus. Optical levitation in air or vacuum does not have this force that reduces the motion of the particle in the trap. For a particle to be trapped in air, it must drift into the focus of the trap with little to no momentum. If the momentum of the particle is too high it will only feel a slight deflection from the force of the laser. Once a particle has been stably levitated, the obvious advantage over other systems that exist are that they are highly isolated from external structures which vastly reduces the decoherence rates of the system. Photoluminescence from optical defects within optically levitated nanodiamond's has been observed [14]. This is especially exciting as information can be encoded into the vibrations and spin structure within the diamonds which can be extracted using the light they emit. In addition these optically levitated objects can be extremely sensitive sensors of forces [15]. For example highly isolated optically trapped particles can be used to investigate the effects of quantum gravity. Unfortunately these systems require extremely high isolation from the environment to reduce the motion of the particle into a quantum ground state. Currently the major limitation for achieving a quantum ground state in these systems is the poor control over the centre of mass motion of the particle. Recently well-established techniques borrowed from atom trapping are being utilised to cool the particles motion in order to approach a centre of mass quantum ground state [16, 17]. However, many of the well-established techniques in atom trapping cannot be used in the optical trapping of nanoparticles due to the lack of distinct energy states in these systems.

1.1.1 Motivation

The negatively charged nitrogen vacancy centre (NV^-) is a nitrogen based optical defect in diamond. It has been thoroughly investigated for its applications in quantum information [18, 19], and nanoscale sensing [20, 21]. It has been shown to be an optically accessible qubit in bulk diamond [22], it displays a long ground state coherence lifetime at room temperature [23] and it can also be used a a source of single photons [24]. As a first step towards trapping nanodiamonds in vacuum, and implementing optomechancal cooling schemes [25, 26], Neukirch et.al. [14] observed the nitrogen vacancy photoluminescence from an optically levitated nanodiamond. In this paper there was an unexpected and unexplained result, which was that the photoluminescence intensity from the NV centre excited by a 532nm pulsed laser was suppressed as the intensity of the 1064nm trapping laser increased. The experimental results obtained from this paper are shown in figure 1.1. Similar results were also obtained in water [27].



Figure 1.1: An increase in photoluminescence spectra observed when the trapping power is reduced, while showing little change in shape. The spectra were measured on the same levitated 100nm nanodiamond containing \sim 500 NV centres [14].

To account for this drop in fluorescence we theorised that the electron population of the NV centre was being trapped in another transition reducing the availability of the electrons to transition in the 637nm state. The transition believed to be the cause of this effect is the optically active 1042nm metastable transition of the NV centres [28]. The 1064nm trapping laser is close enough to this transition to excite the electrons in this metastable state. By taking electrons away from the standard 637nm transition the excited electrons effectively reduce the photoluminescence signal. The probability of keeping the electron in the metastable state is expected to depend on the incident power of the trapping laser, consequently a lower photoluminescence signal for higher powers of the trapping laser as is observed in figure 1.1. It turns out that not only does this effect suppress the photoluminescence, it should also increase the strength of the optical trap. This increase in trapping force is of interest and has not yet been reported. In the context of this I decided to study this system through optical forces.

An optically trapped nanoparticle containing embedded optical defects brings about new regimes of trapping and cooling nanoparticles. Not only does this system allow direct characterisation of the solid state atom properties through force measurements, but by applying well established techniques from atom trapping and cold atom physics to the optically trapped particle, we produce a dramatically stronger trap with the potential to cool the nanoparticle centre of mass motion down past current limits.

These systems could be applied to produce optical traps that trap smaller and smaller nanoparticles, which are of high interest in biological applications [4]. Additionally these effects could produce optically levitated nanoparticles in their quantum ground state at room temperatures providing a platform for studying exotic forces [15, 29, 30] and a variety of non-classical states of light and matter [16, 31].

Alongside the work in this thesis, another part of the group was attempting to investigate quantum trapping on the 637nm transition of the NV centres in nanodiamonds on a customised confocal microscope. The experiment, which is near completion, was designed as a proof of principle of quantum-optical trapping. In this thesis I focus on identifying the trapping forces on the optical defects inside of optically trapped nanoparticles. I develop an experimental setup and procedure to isolate, trap and reliably measure the trapping behaviour of nanoparticles within the optical trap as a platform for investigating the quantum force on optical defects in nanodiamonds. I specifically investigate the quantum force due to the 1042nm metastable transition of the nitrogen vacancy centre and its effect on the harmonic behaviour of the nanodiamonds within the optical trap. The quantum forces due to this metastable state are expected to be much stronger than those arising from the 637nm transition, in addition I hope to provide new insight into the electronic structure of this state.

1.2 Organisation of thesis

The main subject of this thesis is the investigation of optical trapping forces on optical defects within optically trapped nanoparticles.

Chapter 2 contains an extensive review on the optical trapping forces. I introduce the two approaches to optical trapping classical trapping and atom trapping. I develop a qualitative understanding of optical trapping before analysing the individual forces arising from a classical approach for classical trapping and then from a quantum approach for atom trapping.

Chapter 3 contains a review of NV centres in nanodiamonds. I show an overview of the various technologies based on the use of NV centres in diamond. Then I present the physical properties of diamond and then discuss the large variety of optical defects found in diamond. Finally, I focus specifically on the nitrogen vacancy centre in diamond describing the internal energy states and spin structure that this optical defect displays.

Chapter 4 contains the description of the experimental setup. I focus on developing a custom built optical trapping system and developing an imaging system to comprehensibly monitor the motion of particles within the optical trap. I show in detail how the trapping stiffness of the system can be extracted by quantitative measurements of the trapped particles position. I explain the need for high levels of environmental isolation and the methods used to reduce noise within the system. Finally I focus on trapping the nitrogen vacancy centres in diamond and develop the protocols for quantitatively measuring the classical and atomic trapping components.

Chapter 5 contains the conclusions of this work and possible future studies.

2 Optical Trapping

2.1 A Review of Optical Trapping

In 1970, Arthur Ashkin of Bell Labratories calculated that a focused beam of light incident on a microsphere could exert a large force through radiation pressure or conservation of momentum off a reflecting surface [32]. This result motivated a simple experiment [2] to examine such a force on a sample of transparent latex spheres suspended in water. Surprisingly not only did this experiment demonstrate the motion due to laser radiation pressure, but also the existence of a transverse force that results in particle guiding, particle separation, and stable three-dimensional trapping [32]. Optical trapping has found many of its applications in biology. In vitro, optical traps are used to study the properties of a variety of objects such as polymers [33], DNA [34], single molecules [35] and molecular motors [36]. For *in vivo* applications, optical traps are usually employed in a non-quantitative manner to manipulate the relative position of biological objects, such as sorting cells [37] and positioning fluorescent probes [38]. Recently, levitated nanoparticles have been investigated for the development of macroscopic quantum experiments and for high-precision force sensing [16, 17, 26, 39], however the current limitation for these systems is the difficulty of achieving sufficient cooling in order to bring the vibrational motion of the nanoparticle into its quantum ground state. To this end, well-established techniques are being borrowed from atom trapping to reduce the vibrational temperature of optically trapped particles. When optical trapping techniques were applied to the dipole of an optical transition, the main difficulty in providing a stable trap was that the potential wells of the optical traps were only a few kelvin deep. That meant that atoms with a temperature above this value will have enough thermal energy to 'boil' out of the trap. Hence, to provide stable traps, the atoms must be pre cooled to at least this temperature before trapping could occur. Thus as the field of optical trapping of atoms was developed the physics behind laser cooling of atoms was also developed. This brought about a whole new field of research, namely cold atom

physics which provides extreme control over single and collections of atoms. In this regime atoms have been cooled to their lowest quantum states. In this way, coherent superpositions within the trapped atoms have been created providing a test bed for quantum mechanics and manipulation of quantum information [40].

In comparison to atom trapping a trapped, nanoparticle is a massive solid object which is addressed in a different parameter regime. In this regime we can manipulate the centreof-mass motion of the whole system enabling macroscopically distinct superposition states [41]. Additionally the large mass of the solid nanoparticle provides avenues for exceedingly sensitive force sensing [15, 42].

Unfortunately, many of the regimes used for cooling atoms are not applicable to cooling the centre of mass motion of nanoparticles due to the lack of distinct energy levels. By combining the two, atom trapping and classical trapping, into a single system it may be possible to apply a variety of well established laser cooling and cold atom techniques to nanoparticles in order to provide increased control over the trapped nanoparticles, eventually cooling them to their quantum ground state. To achieve this system we must first identify the forces produced from both the classical trapping and those derived from atom trapping techniques. Since the atom trapping forces arise from an approach using quantum formalism, I will be referring to these forces as quantum forces.

2.2 Classical Approach

The most important requirement to achieve a stable optical trap is to create a region of strong intensity gradient. The stronger the gradient field is, the stronger the trap will be. In practice, to produce gradient fields strong enough to enable consistent trapping of particles, a laser beam with a Gaussian transverse profile is tightly focused down to a diffraction limited spot in order to create rapid beam divergence [43]. It is at the centre of this focus that particles can be trapped, and a trapping device of this kind is known as a single beam trap or optical tweezer [44]. Near infrared lasers such as Nd:YAG ($\lambda = 1.064\mu$ m) and Nd:YLF ($\lambda = 1.047\mu$ m) are the most commonly used lasers to manipulate particles in an aqueous medium. These lasers are chosen for three reasons. Firstly, the wavelength is short enough to avoid absorption in water which peaks at 3μ m. Secondly, optical tweezers are often used for biological applications and the biological material is easily damaged by light absorption in the visible region. Finally, laser systems of these types are readily available with extremely high power and stability.

The index of refraction of the particle to be trapped is another consideration to take into account. When the particle has a higher refractive index than the surrounding medium the particle acts as a converging lens to the incoming rays and the net forces pull the particle towards the region of highest intensity. Conversely, if the particles index is smaller, the reverse occurs and the particle will be pushed away from higher intensity regions.

In figure 2.1a and 2.1b a ray optics approach develops a qualitative understanding of the force vectors that arise due to a Gaussian light beam incident on a particle of high refractive index.

Figure 2.1a the focused laser beam refracts off the boundary of the spherical particle. The total refraction off the particle imparts a momentum shift ΔP and hence a force on the



Figure 2.1: Forces on a particle due to a Gaussian light beam. Figure (a) The focused laser beam refracts off the boundary of the spherical particle. The total refraction off the particle imparts a momentum shift ΔP and hence a force on the particle. Due to the particle sitting in the centre of the Gaussian beam symmetry cancels the transverse components and the net force on the particle is backwards towards the focus. (b) If the particle is displaced transversally on the beam axis then the rays entering the particle have uneven amplitudes. In this case the forces due to refraction are uneven and hence there is a net restoring force towards the centre of the trap.

particle. Due to the particle sitting in the centre of the Gaussian beam symmetry cancels the transverse components and the net force on the particle is backwards towards the focus. In figure 2.1b if the particle is displaced transversally on the beam axis then the rays entering the particle have uneven amplitudes. In this case the forces due to refraction are uneven and hence there is a net restoring force towards the centre of the trap.

2.2.1 Geometrical Regime

Trapping was initially performed on particles much larger than the wavelength of light. For such large particles, the trapping beam can be split up into a number of representative rays with powers P and incident angles θ . By considering the forces due to the Fresnel reflections and transmissions of each ray, the total force can be calculated using a summation of the forces due to each ray.

$$F_{scat} = \sum_{i}^{N} \frac{n_m \cdot P_i}{c} \left(1 + R_i \cos(2\theta_{R_i}) - \frac{T_{F_i}^2 (\cos(2\theta_{R_i} - 2\theta_{T_i}) + R_i \cos(2\theta_{R_i}))}{1 + R_i^2 + 2R_i \cos(2\theta_{T_i})} \right)$$
(2.1)

$$F_{grad} = \sum_{i}^{N} \frac{n_m \cdot P_i}{c} \left(1 + R_i \sin(2\theta_{R_i}) - \frac{T_{F_i}^2 (\sin(2\theta_{R_i} - 2\theta_{T_i}) + R_i \cos(2\theta_{R_i}))}{1 + R_i^2 + 2R_i \cos(2\theta_{T_i})} \right), \quad (2.2)$$

where R and T_F are the Fresnel coefficients, θ_R and θ_T are the angles for reflection and transmission of the incident rays. This regime gives an understanding of particle trapping, however it is only valid for particles where the particle radius is much larger than the wavelength $(a \gg \lambda)$. Moreover for most optical trap, and all those considered in this thesis, the particle is much smaller and hence the Rayleigh regime must be considered to describe the trapping behaviour.

2.2.2 Rayleigh Regime

In the Rayleigh regime case the beam profile is very important for the trapping behaviour and hence must be described before we look closely into the trapping forces. The conventional beam profile used in optical traps and those used in this thesis are the Gaussian beam profile, the geometry of which is shown in figure 2.2.



Figure 2.2: Geometry of a particle of radius a at a position $\mathbf{r} = (x, y, z)$ in a Gaussian beam with a beam waist W_0 .

Using a paraxial Gaussian beam description with linear polarisation, the electric-field vector at the position \boldsymbol{r} in terms of complex amplitude $\mathcal{E}(\boldsymbol{r},t)$ is given by [45]

$$\begin{aligned} \mathcal{E}(\mathbf{r},t) &= \hat{x} \frac{1}{2} \sqrt{\frac{4P}{\pi W_0^2 n_m \epsilon_0 c}} \frac{ikW_0^2}{ikW_0^2 + 2z} \exp\left[-\frac{(kW_0)^2 (x^2 + y^2)}{(kW_0^2)^2 + (2z)^2}\right] e^{-ikz} \\ &\times \exp\left[-i\frac{2kz(x^2 + y^2)}{(kW_0^2)^2 + (2z)^2}\right] e^{-i\omega t}. \\ &= \hat{x} \frac{\mathbf{E}_0(\mathbf{r})}{2} e^{i\phi(\mathbf{r})} e^{-i\omega t} \end{aligned}$$
(2.3)

where \hat{x} is the unit vector in the polarisation direction, P is the beam power given by $P = \pi W_0^2 n_m \epsilon_0 c E_0^2/4$, W_0 is the beam width, ω is the trapping frequency, k is the wave number in the medium given by $k = n_m \omega/c$ and E_0 is the electric field strength. The associated magnetic field vector under this approximation is given by

$$\boldsymbol{H}(\boldsymbol{r},t) = \hat{z} \times \frac{\mathcal{E}(\boldsymbol{r},t)}{Z_0} \simeq \hat{y} n_m \epsilon_0 c \mathcal{E}(\boldsymbol{r},t) = \hat{y} H(\boldsymbol{r},t), \qquad (2.4)$$

where, $Z_0 = \sqrt{\mu_m/\epsilon_m} \simeq 1/(n_m\epsilon_0 c)$ is the intrinsic impedance of the medium, $c = 1/\sqrt{\epsilon_0\mu_0}$ is the speed of light and ϵ_0 and μ_0 are the vacuum permittivity and permeability, respectively. The Poynting vector, which describes the instantaneous energy flux crossing a unit area per unit time is thus given by

$$\boldsymbol{S}(\boldsymbol{r},t) \equiv \mathcal{E}(\boldsymbol{r},t) \times \boldsymbol{H}(\boldsymbol{r},t)$$
(2.5)

The beam intensity, defined as the time-averaged version of the Poynting vector is then given by

$$I(\mathbf{r}) \equiv \|\langle \mathbf{S}(\mathbf{r},t) \rangle_T\|$$
(2.6)

$$= \|\tilde{z}\frac{n_m\epsilon_0 c}{2}|\boldsymbol{E}_0(\boldsymbol{r})|^2\|$$
(2.7)

$$= \left(\frac{2P}{\pi W_0^2}\right) \frac{1}{1 + (2\tilde{z})^2} \exp\left[-\frac{2(\tilde{x}^2 + \tilde{y}^2)}{1 + (2\tilde{z})^2}\right].$$
 (2.8)

in which \tilde{x}, \tilde{y} and \tilde{z} are the normalised spacial co-ordinates given by $(\tilde{x}, \tilde{y}, \tilde{z}) = (x/\omega_0, y/\omega_0, z/k\omega_0^2)$. These descriptions use the paraxial approximations of Maxwell's equations to the scalar wave equation of the Gaussian beam, valid when $W_0 \gg \lambda$, [46, 47].

For particles where the wavelength of light is much greater than the size of the particle $(r \ll \lambda)$, the instantaneous electric field acting across the particle can be taken to be uniform. In this case the electrostatics formulae can be applied. Similar to the ray optic regime, the force exerted on the particle in the Rayleigh regime can be described by two components acting on the dipole, the scattering force and the gradient force.

2.2.3 General expression of the radiative forces

To develop the motion of the particles within an optical trap we must first look at the radiative forces on the particles caused by an electric field acting on the dipole moment of the particle. In this case an atom can be considered as a very small particle, the difference from a classical particle arises in the nature of the dipole moment. The dipole moment of a classical particle is an induced dipole caused by the electric field, whereas on the other hand, the atomic dipole moment is given by the inherent electronic structure. The optical force on the expectation value of the dipole moment of the particle $\langle \hat{D}_{\epsilon} \rangle$ at position r_0 is:

$$\boldsymbol{F} = \langle \boldsymbol{\hat{D}}_{\epsilon} \rangle \nabla \boldsymbol{E}(\boldsymbol{r}, t)_{\boldsymbol{r_0}}.$$
(2.9)

The particle undergoes forced oscillations at the frequency ω of the field, and hence we can write

$$\langle \hat{\boldsymbol{D}}_{\epsilon} \rangle = \epsilon_0 \alpha \mathcal{E} + c.c,$$
 (2.10)

where, α is the polarisability of the particle. The polarisability is a complex number,

$$\alpha = \alpha' + i\alpha'' \tag{2.11}$$

which depends on the wavelength and the field amplitude, and has the dimensions of length cubed.

We now assume that the internal state of the particle reaches a steady state at a point r_0 under the effect of the field,

$$\boldsymbol{E}(\boldsymbol{r},t) = \boldsymbol{\mathcal{E}}(\boldsymbol{r},t) + \boldsymbol{\mathcal{E}}^*(\boldsymbol{r},t), \qquad (2.12)$$

recalling

$$\mathcal{E}(\mathbf{r},t) = \hat{x} \frac{\mathbf{E}_0(\mathbf{r})}{2} e^{i\phi(\mathbf{r})} e^{-i\omega t}.$$
(2.13)

And now substituting equation (2.10) into (2.9), we obtain four force terms,

$$\boldsymbol{F} = \epsilon_0 \alpha \mathcal{E} \nabla \mathcal{E}_{\boldsymbol{r}_0} + \epsilon_0 \alpha^* \mathcal{E}^* \nabla \mathcal{E}^*_{\boldsymbol{r}_0}$$

$$+ \epsilon_0 \alpha \mathcal{E} \nabla \mathcal{E}^*_{\boldsymbol{r}_0} + \epsilon_0 \alpha^* \mathcal{E}^* \nabla \mathcal{E}_{\boldsymbol{r}_0}$$
(2.14)

$$=\epsilon_0 \alpha' \frac{\boldsymbol{E_0}(\boldsymbol{r})}{2} \nabla \boldsymbol{E_0}(\boldsymbol{r})_{\boldsymbol{r}_0} - \epsilon_0 \alpha'' \frac{\boldsymbol{E_0}^2(\boldsymbol{r})}{2} \nabla \phi(\boldsymbol{r})_{\boldsymbol{r}_0}.$$
(2.15)

The two first terms oscillate at 2ω and give rise to no effects when averaged over time. The two remaining terms do not oscillate at all and thus give rise to a force. The radiative force thus comprises two contributions. The first of these relates to the real part α' of the polarisability and depends on the gradient field of the electromagnetic wave. This corresponds to the dipole force, or the gradient force. The other contribution relates to the imaginary part α'' of the polarisability and depends on the gradient of the phase of the wave. This force corresponds to the resonant scattering force which is related to the absorption of the photons. One needs to be careful distinguishing this resonant scattering force due to absorption and the scattering force due to the Rayleigh scattering off the particle in classical trapping.

Gradient force

For a small particle in an aqueous medium,

$$\alpha = \alpha' + i \cdot \alpha'' = 4\pi n_m^2 a^3 \left(\frac{m^2 - 1}{m^2 + 2}\right)$$
(2.16)

is the relative complex polarisability of the particle in a surrounding medium, where, n_p and n_m is the refractive index of the particle and medium respectively and $m = n_p/n_m$ is the relative refractive index of the particle. Thus by substituting equation (2.16) into equation (2.15) we have,

$$\boldsymbol{F}_{grad}(\boldsymbol{r}) = \mathbb{R}\left(4\pi n_m^2 \epsilon_0 a^3 \left(\frac{m^2 - 1}{m^2 + 2}\right)\right) \frac{\boldsymbol{E}_0(\boldsymbol{r})}{2} \nabla \boldsymbol{E}_0(\boldsymbol{r})_{\boldsymbol{r}_0}.$$
(2.17)

Target particles for optical trapping are almost always transparent dielectric spheres. In this case, the complex component of the polarisability is zero ($\alpha'' = 0$). Therefore the scattering force due to the absorption of the particle can be neglected. Using the vector identity $\nabla E^2 = 2(E \cdot \nabla)E + 2E \times (\nabla \times E)$ with $\nabla \times E = 0$ from Maxwells equations we have,

$$\boldsymbol{F}_{grad}(\boldsymbol{r}) = 4\pi n_m^2 \epsilon_0 a^3 \left(\frac{m^2 - 1}{m^2 + 2}\right) \frac{1}{2} \nabla \langle \boldsymbol{E}^2(\boldsymbol{r}, t) \rangle_T$$
(2.18)

$$=\pi n_m^2 \epsilon_0 a^3 \left(\frac{m^2 - 1}{m^2 + 2}\right) \frac{1}{2} \nabla |\boldsymbol{E}(\boldsymbol{r}, t)|^2$$
(2.19)

$$= \frac{2\pi n_m^2 a^3}{c} \left(\frac{m^2 - 1}{m^2 + 2}\right) \frac{1}{2} \nabla I(\boldsymbol{r}).$$
(2.20)

By substituting the beam intensity from equation (2.8) the gradient force can expressed in terms of each of its three components,

$$\boldsymbol{F}_{grad,x}(\boldsymbol{r}) = \hat{x} \frac{2\pi n_m^2 a^3}{c} \left(\frac{m^2 - 1}{m^2 + 2}\right) \frac{4\hat{x}/W_0}{1 + (2\hat{z})^2} \\ \times \left(\frac{P}{\pi W_0^2}\right) \frac{1}{1 + (2\hat{z})^2} \exp\left[-\frac{2(\hat{x}^2 + \hat{y}^2)}{1 + (2\hat{z})^2}\right]$$
(2.21)

$$\boldsymbol{F}_{grad,y}(\boldsymbol{r}) = \hat{y} \frac{2\pi n_m a}{c} \left(\frac{m-1}{m^2+2}\right) \frac{4g/W_0}{1+(2\hat{z})^2} \times \begin{pmatrix} P \\ 0 \end{pmatrix} \frac{1}{1-(2\hat{x}^2+\hat{y}^2)}$$
(2.22)

$$\times \left(\frac{1}{\pi W_0^2}\right) \frac{1}{1 + (2\hat{z})^2} \exp\left[-\frac{1}{1 + (2\hat{z})^2}\right]$$

$$F_{grad,z}(\mathbf{r}) = \hat{z} \frac{2\pi n_m^2 a^3}{c} \left(\frac{m^2 - 1}{m^2 + 2}\right) \frac{8\hat{z}/(kW_0^2)}{1 + (2\hat{z})^2}$$

$$\left[-2(\hat{x}^2 + \hat{x}^2)\right] \left(-2R_0\right) = 1 \quad [-2(\hat{x}^2 + \hat{x}^2)]$$
(2.22)

$$\times \left[1 - \frac{2(\hat{x}^2 + \hat{y}^2)}{1 + (2\hat{z})^2}\right] \left(\frac{2P}{\pi W_0^2}\right) \frac{1}{1 + (2\hat{z})^2} \exp\left[-\frac{2(\hat{x}^2 + \hat{y}^2)}{1 + (2\hat{z})^2}\right]$$
(2.23)

In contrast to the scattering force of equation (2.30), the gradient force consists of three components which act as restoring forces directed towards the centre of the focused laser beam for particles of high refractive index. For particles of lower refractive index than the medium, the particles are pushed away from the focus. In fact this can be generalised for all beam shapes: particles of high refractive index are attracted to regions of high intensity, whereas particles of lower relative refractive index are repelled from areas of high refractive intensity, and can even be trapped in minima of local intensity.

Another feature of the gradient force is that by integrating equation (2.20) we see that the dipole force acts as if it derives from a potential.

$$\boldsymbol{F}_{grad}(\boldsymbol{r}) = -\nabla U_{cl}(\boldsymbol{r}) \tag{2.24}$$

where

$$U_{cl}(\boldsymbol{r}) = \frac{2\pi n_m a^3}{c} \left(\frac{m^2 - 1}{m^2 + 2}\right) \frac{2P}{\pi W_0^2}.$$
(2.25)

To achieve trapping the depth of this potential well should be much larger than the average kinetic energy of the particle generated by Brownian motion $(k_B T)$.

Classical scattering force

The derivation of the classical scattering force does not arise from equation (2.15) but arises as a consequence of photons having momentum. The force of the scattering photons acts in the direction of the propagation of the light and is dependent on the light intensity rather than the intensity gradient. The momentum of a single photon of energy J is:

$$\boldsymbol{p} = \hbar \boldsymbol{k} = \frac{\boldsymbol{J} n_m}{c} \tag{2.26}$$

A beam of incident photons can be scattered by a particle resulting in two impulses. The force on the particle is equal to the difference between the momentum of the input beam and the scattered field from the particle. The scattered field, since the particle is in the Rayleigh regime is isotropic in all directions. Hence, the impulse due to the scattered light has no preferable direction and results in no net force. The change in momentum, or force, of a particle can be calculated by considering the photon flux impinging on an object under the conservation of momentum:

$$\boldsymbol{F}_{scat} = \hat{z} \frac{n_m \sigma}{c} \langle \mathbf{S} \rangle_T \tag{2.27}$$

$$= \hat{z}\left(\frac{n_m}{c}\right)\sigma I(\boldsymbol{r}) \tag{2.28}$$

where n_m is the refractive index of the particle, $\langle \mathbf{S} \rangle$ is the time averaged Poynting vector, c is the speed of light and σ is the scattering cross section for the radiation pressure of the particle. In the case of a small dielectric particle in the Rayleigh regime where the light scatters isotropically, the scattering cross section σ is given by [48],

$$\sigma = \frac{8}{3}\pi \left(ka\right)^4 a^2 \left(\frac{m^2 - 1}{m^2 + 2}\right)^2.$$
 (2.29)

By substituting equations (2.8) and (2.29) into equation (2.28), the scattering force is given in terms of the intensity distribution of the beam by

$$\hat{F}_{scat}(\hat{r}) = \hat{z}\frac{8}{3}\pi \left(ka\right)^4 a^2 \left(\frac{m^2 - 1}{m^2 + 2}\right)^2 \times \left(\frac{2P}{\pi\omega_0^2}\right) \frac{1}{1 + (2\hat{z})^2} \exp\left[-\frac{2(\hat{x}^2 + \hat{y}^2)}{1 + (2\hat{z})^2}\right].$$
(2.30)

This expression shows that this trapping force is highly dependent on the light intensity, the wavelength and the size of the particle to be trapped.

2.3 Quantum Forces

The conceptual understanding of atomic forces are the same as those understood from classical trapping. The atoms can be considered small particles trapped in the Rayleigh regime, the only difference is the polarisability of the atom produces strong resonant properties around atomic transitions. In the case of classical trapping an intuitive understanding arises from the refraction of rays of light through a spherical particle, in the atom trapping case photons are absorbed by a two level system and then emitted isotropically. This absorption and isotropic emission produces the forces in equation (2.15). In this case the two separate forces, a dipole type force and a resonant scattering type force arise from the polarisability having both a real and an imaginary part.



2.3.1 Polarisability of a two level atom

Figure 2.3: Polarisability of a 637nm two level transition, with $2\pi \times 20$ MHz transition linewidth at the centre of an optical trap with 25mW of power at the focal spot of a 1.2NA objective. The imaginary part of the polarisability has been multiplied $\times 100000$ to fit on the same scale as the real part of the polarisability.

The polarisability of a two-level atom with zero velocity is

$$\alpha = \frac{d^2}{\epsilon_0 \hbar} \frac{\omega_0 - \omega + i\Gamma/2}{(\omega_0 - \omega)^2 + \frac{1}{2} \left(\frac{dE_0}{\hbar}\right)^2 + \frac{\Gamma^2}{4}},\tag{2.31}$$

where, d is the dipole moment of the transition, Γ is the transition linewidth, ω_0 is the transition frequency, ω is the trapping laser frequency and E_0 is the electric field strength. Rewriting this as a linear polarisability term, independent of the wave intensity, with a saturation term $(1 + s)^{-1}$ which tends to zero at high intensities:

$$\alpha = \frac{d^2}{\epsilon_0 \hbar} \frac{\omega_0 - \omega + i\Gamma/2}{(\omega_0 - \omega)^2 + \frac{\Gamma^2}{4}} \frac{1}{1+s}.$$
(2.32)

The saturation parameter s is given by

$$s = \frac{1}{2} \left(\frac{d\mathbf{E_0}}{\hbar} \right)^2 \frac{1}{(\omega_0 - \omega)^2 + \frac{\Gamma^2}{4}} = \frac{I}{I_{sat}} \frac{1}{1 + 4\left(\frac{\omega - \omega_0}{\Gamma}\right)},$$
(2.33)

with

$$I_{sat} = \frac{1}{I\Gamma^2} \left(\frac{d\boldsymbol{E_0}}{\hbar}\right)^2.$$
(2.34)

We can see from equation (2.31) the polarisability of the two-level atom shows resonant behaviour about the transition frequency $\omega \sim \omega_0$. Note also that if the velocity \boldsymbol{v} of the atom is non-zero, the Doppler effect should be taken into account in the polarisability, by replacing ω by $\omega - \boldsymbol{k} \cdot \boldsymbol{v}$. This non zero velocity plays a very important role in Doppler cooling of atoms.

2.3.2 Dipole Force

To observe a dipole force it is essential to have a gradient profile across E_0 . For example a common beam profile is the Gaussian profile,

$$\boldsymbol{E}_{0}(x,y) = A_{0}e^{-\frac{x^{2}+y^{2}}{\omega_{0}^{2}}},$$
(2.35)

propagating in the z direction, The electromagnetic field has a complex amplitude

$$\mathcal{E}(\mathbf{r},t) = \frac{A_0}{2} e^{\frac{-x^2 + y^2}{\omega_0^2}} e^{ikz} e^{-i\omega t}.$$
(2.36)

Substituting into equation (2.15) the first dipole force term now gives a non zero contribution:

$$F_2 = \epsilon_0 \alpha' \frac{\boldsymbol{E_0}}{2} \nabla \boldsymbol{E_0} = \epsilon_o \frac{\alpha'}{4} \nabla (\boldsymbol{E_0}^2).$$
(2.37)

Using the real part of the polarisability of a two level atom gives

$$\boldsymbol{F}_{2} = \frac{\hbar(\omega_{0} - \omega)}{2} \frac{\nabla(\frac{1}{2} \left(\frac{d\boldsymbol{E}_{0}}{\hbar}\right)^{2})}{(\omega_{0} - \omega)^{2} + \frac{1}{2} \left(\frac{d\boldsymbol{E}_{0}}{\hbar}\right)^{2} + \frac{\Gamma^{2}}{4}}.$$
(2.38)

We can see now that the dipole force varies directly with the gradient of the light intensity. There is also interesting behaviour about the atomic transition ω_0 . For a blue detuning $(\omega > \omega_0)$ the atoms are repelled from regions of high intensity, whereas for red detuning $(\omega < \omega_0)$ the atoms are attracted towards such regions.

By integrating equation (2.38) we see that the dipole force acts as if it derives from a potential. The choice of integration constant is chosen to give a zero potential outside the laser beam, so that,

$$\boldsymbol{F}_2 = -\nabla U_q(\boldsymbol{r}),\tag{2.39}$$

with

$$U_q(\nabla) = \frac{\hbar(\omega - \omega_0)}{2} \ln\left[1 + \frac{1}{2} \left(\frac{d\mathbf{E_0}}{\hbar}\right)^2 \frac{1}{(\omega_0 - \omega)^2 + \frac{\Gamma^2}{4}}\right]$$
(2.40)

$$=\frac{\hbar(\omega-\omega_0)}{2}\ln\left[1+s(\boldsymbol{r})\right]$$
(2.41)

$$=\frac{\hbar(\omega-\omega_0)}{2}\ln\left[1+\frac{I}{I_{sat}}\frac{1}{1+4\left(\frac{\omega-\omega_0}{\Gamma}\right)}\right].$$
(2.42)

To compare with the classical potential for the dipole force given in equation (2.25), we substitute in the relation for the electric field amplitude at the trap centre into equation (2.40), which gives,

$$U_q = \frac{\hbar(\omega - \omega_0)}{2} \ln\left(1 + \frac{1}{2}\frac{d^2}{\hbar^2}\frac{4P}{\pi W_0^2 n_m \epsilon_0 c}\frac{1}{(\omega - \omega_0)^2 + \frac{\Gamma^2}{4}}\right).$$
 (2.43)

This potential presents a very strong dependence with the wavelength. This also suggests that it may be possible to trap atoms at the focal point of a laser beam, if the frequency is red detuned ($\omega < \omega_0$). The potential has a minimum in both the x and y dimensions of typical width ω_0 , and also has a minimum in the z direction of typical width $z_R = \pi \omega_0^2 / \lambda$, where z_R is the Rayleigh length. The atoms will be trapped within the potential well provided that their kinetic energy is less than the depth of well. Due to this effect in practice it is difficult to produce potential wells able to trap atoms with temperatures above a few milikelvins.



Figure 2.4: Optical potential depth for a single 637nm transition, with $2\pi \times 20$ MHz transition linewidth at the centre of an optical trap with 25mW of power at the focal spot of a 1.2NA objective. The particle depth is expressed in fraction of average kinetic energy (k_BT at T = 300K).

2.3.3 Resonance scattering Force

To isolate the force due to the resonance scattering we consider a travelling plane wave,

$$\mathcal{E}(\boldsymbol{r},t) = \frac{A_0}{2} e^{i\boldsymbol{k}\cdot\boldsymbol{r}} e^{-i\omega t}, \qquad (2.44)$$

with constant amplitude E_0 and wave-vector k, where $|k| = k = \omega/c$. As can be seen from equation (2.15) the only radiative force is the resonance scattering since the gradient of the

amplitude of the plane wave is zero $(\nabla E_0(\mathbf{r}) = 0)$.

$$\boldsymbol{F}_{1} = \epsilon_{0} \alpha'' \frac{\boldsymbol{E_{0}}^{2}}{2} \nabla(\boldsymbol{k} \cdot \boldsymbol{r}) = \epsilon_{0} \alpha'' \frac{\boldsymbol{E_{0}}^{2}}{2} \boldsymbol{k}.$$
(2.45)

The force lies along the direction of propagation k of the wave. Now substituting the imaginary part of the polarisability of a two-level atom from equation (2.31) we have,

$$\boldsymbol{F}_{1} = \frac{d^{2}}{2\hbar} \frac{\Gamma}{(\omega_{0} - \omega)^{2} + \frac{1}{2} \left(\frac{d\boldsymbol{E}_{0}}{\hbar}\right)^{2} + \frac{\Gamma^{2}}{4}} \frac{\boldsymbol{E}_{0}^{2}}{2} \boldsymbol{k}.$$
(2.46)

By rearranging, and then using the saturation parameter s given in equation (2.33), F_1 becomes

$$\boldsymbol{F}_{1} = \hbar \boldsymbol{k} \frac{\Gamma}{2} \frac{1}{2} \left(\frac{d\boldsymbol{E}_{0}}{\hbar} \right)^{2} \frac{1}{(\omega_{0} - \omega)^{2} + \frac{1}{2} \left(\frac{d\boldsymbol{E}_{0}}{\hbar} \right)^{2} + \frac{\Gamma^{2}}{4}} = \hbar \boldsymbol{k} \frac{\Gamma}{2} \frac{s}{1+s}.$$
 (2.47)

Since s behaves resonantly we also observe that the resonant scattering has a resonance around the atomic frequency ω_0 , in this case a Lorentzian resonance. This resonance is narrow, with the width of the resonance on the same order as the width Γ of the atomic line.

3

Nitrogen Vacancy Centres in Diamond

Known for thousands of years, diamond has not only been used as an engraving tool, but also as a prized gemstone. The popularity of diamond grew rapidly in the 19^{th} century when techniques improved to cut and shape the material. In fact the popularity of diamonds was transformed by the marketing campaign of De Beers', who owned and still owns the majority of the world's diamond supplies. De Beers' marketing campaign focused on advertising the product rather than the brand and can be argued to be one of the greatest advertising campaigns, the result of which made diamonds a true icon in the eyes of the public. These diamonds are natural diamonds which are not perfect but are found in many different types and colours, each with different optical and physical properties. These imperfections are desirable in jewellery producing their vibrant sparkles and colours, however these imperfections are usually undesirable for technological purposes on a commercial scale. Commercial applications of diamonds have reached many fields of science. This is because diamond comprises of extreme properties, which combined are unparalleled by any other single material. Whether it be mechanical, electrical, thermal or optical the varying and extreme values have pushed diamond based applications into a diverse range of fields, such as mechanical machining, high-power high frequency electronics, high power non-linear optics, and many more [49]. Recently advancements in diamond synthesis, now provides us with dramatically improved diamond samples with specified properties of our choosing. These new techniques, not only give the science community the possibility of tuning the characteristics of diamonds, but now diamonds can be grown with a wide variety of desirable defects, referred to as optical defects or colour centres, which are responsible for the all the colours and vibrancy natural diamond displays. Beyond the beauty of the colour, many of these defects made from atomic inclusions show interesting spin and optical properties which have produced promising applications in the science community.

Of all the optical defects in diamond, the NV centre stands out as the most appealing for scientific purposes and is prominent in at least three fields of research: high-resolution magnetometry, biomedicine, and quantum information technology. This is only a short overview, however there are a number of reviews that deal specifically with the abundance of NV centre applications [18, 50–53].

NV centres have found promising applications in detecting single spins and weak magnetic fields. The external magnetic fields acting on a single NV centre can measurably alter the florescence. The detection of the modified fluorescence of the NV can be used to probe the surrounding fields. This technique has even been able to detect electron spin and nuclear spins in ambient conditions [54, 55]. NV centres for high resolution megnetometry have been used in encouraging applications for read out of single spin based magnetic memories and for high-resolution imaging techniques [20, 21, 56, 57]

Recently research into colour centres in diamond for biological and medical purposes has been quite active. Nanodiamonds containing fluorescent centres have several attractive properties. They are biocompatible and non-toxic due to their inert carbon structure. They can also be excited by a laser through tissue and the resulting emission from the defect can also be capable of penetrating tissues. These characteristics are difficult to be found together as traditional fluorescent molecules used in biology either blink or photobleach. Hence fluorescent nanodiamonds are being developed as new types of probes for bio-labelling and as drug delivery vehicles [58–61].

NV defects in diamond are also being investigated for their implementation of solid state quantum technologies. One avenue is to use NV centres in diamond as addressable and easy to manipulate quantum states (qubits), even in room temperature conditions [62]. There are several properties that provide NV centres as an excellent candidate for use in quantum information. Firstly it is a single photon emitter which shows high oscillator strength for the dipole transition between the ground and the first optically excited state. The spin state can be optically initialised, coherently manipulated and read out with optical and microwave excitation and lastly, the NV centre has shown appealing long coherence lifetimes at room temperature [18, 51, 52, 63]. Although it is a promising element for building quantum devices, there are still limitations that prevent these devices from true realisation. For example, the NV centre continues to show a suppression of emission spectra in optical trapping experiments [14].

It is generally accepted that there is a real potential for colour centres in diamond, but limitations in understanding the internal electronic structure present obstacles that prevent the quality and accuracy required for the implementation of quantum devices.

3.1 Physical properties of diamond

Diamond is and allotrope of carbon, where each atom sits in a periodic structure with strong covalent bonds with its four nearest neighbours arranged at the corners of a regular tetrahedron as shown in figure 3.1. Each covalent bond in the lattice is an identical bond with a length of 0.154nm [64], eight atoms are needed to form the unit cubic cell which has a length of 0.365nm [64]. Due to its tetrahedral structure with identical covalent bonds diamond is the hardest known natural material. Diamond has a Vickers hardness of 115GPa, whereas for comparison iron has a hardness value of 0.6GPa [65]. Due to its extremely high hardness, since its discovery it has been used extensively for cutting, grinding and polishing.

But unlike other hard materials diamond also has a high thermal conductivity and a low friction coefficient. These additional factors together with the hardness are what provides diamond to be so effective for its varied mechanical applications.



Figure 3.1: Diamond lattice cubic cells. **a)** Position of the carbon atoms in a unit cell projected on a cube face. The fractions denote the height above the base in units of a cube edge length. **b)** Three dimensional view of the diamond cubic cell with black sticks representing covalent bonds [66].

3.2 Optical properties of diamond

Pure diamond is optically transparent unlike those that are found in nature which exhibit many impurities and defects which makes those diamonds valuable as prized gemstones. Diamond is highly transparent from the near UV all the way through to the far infrared that traditionally made it useful for optical windows in infrared instrumentation. In addition to the transparency the refractive index is a very important property of a material as so many other optical properties rely on the refractive index of a material. Pure diamond displays a relatively high refractive index which may vary slightly according to the type of diamond. The dependence of the wavelength on the refractive index are shown in figure 3.2.

The critical angle of light, which is the limiting angle where total internal reflection occurs, depends on the difference of refractive index between two materials. For diamond in air the critical angle is 24.6° and for diamond in water it is 33.6° . Reflectance also depends on the refractive index difference between two materials as well as depending on the incident angle on the diamond. In air diamond reflects 17% of the light hitting at an angle of 90° and 27% of light hitting at 30°. The corresponding reflectance values for window glass at these angles are 4% and 17% respectively [67]. Another optical property of some interest is the birefringence of a material, although due to the cubic structure of diamond it does not exhibit birefringence. Briefly mentioned earlier, the variety of colours and varied



Figure 3.2: Plot of the dependence of the wavelength on the refractive index of diamond. It can be observed that the refractive index remains fairly constant (~ 2.4) from the visible to the far infrared. In comparison water has a refractive index of ~ 1.33 and fused quartz has a refractive index of ~ 1.46 .

optical properties of diamond arise from internal defects in the diamond structure and will be discussed in detail in the following section.

3.3 Optical defects in diamond

Defects in diamond are responsible for the colourful variety of natural diamonds found throughout the world. These defects give rise to many interesting properties, of particular interest are the optical properties which we will soon explore. Defects in diamond can be caused by internal irregularities in the structure of the diamond lattice itself. Defects of this kind are called intrinsic defects and can be the result of extra or missing carbon atoms as well as lattice sites with broken bonds. The other cause of defects in diamond are when an atomic inclusion enters the diamond matrix. Defects of this type are called extrinsic defects and are the result of carbon atoms being replaced by the atom inclusion, or the atom sitting in place between lattice sites. A combination of intrinsic and extrinsic defects can produce an optical defect, a system with its own specific energy levels separate to that of the diamond structure. Many atomic inclusion impurities are known to form optically active defects in diamond including: H, He, Li, B, N, O, Ne, P, Si, As, Ti, Cr, Ni, Co, Zn, Zr, Ag, W, Xe and
Tl [66, 68–70]. In fact diamond has been found to contain over 500 optically active defects over the range of 170nm to 20μ m [68].

The most notable diamond defects arise from nitrogen related defects as they are the most common impurity to occur in diamond due to nitrogen's abundance in the atmosphere. Of all the structures that nitrogen can form inside diamond, the most studied is the nitrogen-vacancy centre or simply NV, shown in figure 3.3.



Figure 3.3: NV structure in the diamond lattice and photoluminescence structure. (a) The NV centre has a principal axis of symmetry in the crystallographic [111] direction. The nitrogen atom is shown in black, carbon atoms are in grey, and the vacancy in white with no bonds (black sticks) [66]. (b) Fluorescence emission spectra of single NV centres at room temperature and liquid helium temperatures. The excitation wavelength was 514nm [52].

The NV centre is an extrinsic diamond crystallographic defect incorporated in the tetrahedral diamond structure of four carbon atoms where two adjacent sites are altered. One carbon atom is replaced by a nitrogen atom and the other is replaced by a vacant space (vacancy). Being in group V of the periodic table the nitrogen atom has five valence electrons. Three of the electrons bond covalently with the three nearest neighbours the remaining two occupy the dangling bond in the direction of the vacancy. The vacancy picks up an additional three dangling bonds from its three nearest carbon neighbours providing five unsatisfied active bonding elements. It has been estimated that in the NV complex ~ 72% of the charge density is at the three nearest carbon neighbour atoms surrounding the vacancy, only ~ 0.2% is at the nitrogen and approximately ~ 28% is spread over the lattice [71, 72].

The NV centre can exist in two separate charge states which depends mainly on the local lattice environment. The presence of close surrounding impurities acting as electron acceptors or donors cause the NV to be in either its neutral NV⁰ state or in its negatively charged NV⁻ state [73, 74]. NV⁰ and NV⁻ can be found occurring within the same diamond [74]. It has also been observed that the same NV defect can undergo photo-chromic switching between the two charge states. The mechanisms that provide the photo-conversion can be either direct, ionisation of a negatively charged NV⁻ or, indirectly, the ionisation of nitrogen donors surrounding the NV centre in the crystal [75, 76]. The two charge states are clearly

identified separately due to their spectrally different characteristic optical transitions [68]. The NV⁻ has a strong optical transition with a zero phonon line (ZPL) i.e. the pure electronic transition, at 637nm (1.945 eV) again seen in figure 3.3b. The ZPL is associated with an electric dipole transition between isolated states of spatial symmetry ground state ${}^{3}A_{2}$ and excited state ${}^{3}E$ [77]. The ZPL emission is accompanied by a large phonon side band. At room temperature there is enough thermal energy to excite many phonons and the emission spectra is spread out over longer wavelengths. It is clear in the emission spectra in figure 3.3b, that the area contained within the sharp ZPL actually represents only a tiny fraction of the total emission. This fraction can be increased by reducing the phonon side band through cooling the NV centres down to low temperatures. However even at low temperatures the ZPL signal is still only a small fraction of the emission spectra, for example at liquid helium temperature (10K) the ZPL only represents ~3% of the total emission [78]. One should also note that the relative position of the ZPL and phonon side band is reversed for absorption.

3.4 Energy States

The electronic structure of the NV centre within diamond is an interesting feature and substantial efforts have been made to completely resolve the structure. Here we focus on the negatively charged NV⁻ centre, since it turns out to have more interesting physics in terms of spin, magnetic and optical properties. A detailed description of the NV⁻ energy level scheme based on theoretical considerations and experiments have been given [79–81]. To present the spin and optical properties of the NV centre in diamond, a simplified energy level scheme of the centre is considered and shown in figure 3.4.

The NV⁻ centre is treated as a four-level system, with two triplet states, ³A and ³E, and two singlet states, ¹A and ¹E. The separation in energy between the triplet state and the optically excited state ³E is 1.945eV (ZPL at 637nm). Both triplet states are split by the spin sublevels of the spin quantum number $m_s = 0$ and $m_s = \pm 1$. For the ground state the zero-field splitting between $m_s = 0$ and $m_s = \pm 1$ has been measured to be $D_{gs} = 2.88GHz$, which means this transition is accessible in via microwave excitation. The two ground states $m_s = \pm 1$ are degenerate due to symmetry reasons however a local change in symmetry due to lattice irregularities, strain or external fields can alter the degeneracy, making the sub levels distinguishable.

There was constant debate in the literature about the structure the metastable singlet state. Symmetry considerations predicted two singlet states, a short lived ${}^{1}E$ and a longer lived metastable state ${}^{1}A$ [79]. However other theoretical predictions arose contradicting this assignment [83, 84]. In the literature these states are still usually described as a single metastable state, however the state has been experimentally verified as two singlet states with an optically active transition at 1.190eV [85]. The short lived ${}^{1}E$ has a lifetime of ≤ 1 ns and the longer lived metastable state ${}^{1}A$ has a lifetime of $\sim 150 - 450$ ns [85]. Although this state has been observed it has still not been fully characterised. For example the electron transitions from the $m_s = \pm 1$ sublevel of the excited triplet state into the excited singlet state into the metastable ground state (${}^{1}A$) the electron transitions predominately from the excited singlet state into the $m_s = 0$ state of the ground triplet state. In this process there is an anti-crossing in



Figure 3.4: Scheme of the NV⁻ electronic structure. The ground ³A and the excited ³E states are triplets and have a separation energy of 1.945eV [82]. The singlet state transition has a separation energy of 1.190eV [28] and can only be optically addressed when the centre is being excited at 1.945eV. The ground state zero field splitting is $D_{gs} = 2.88$ GHz. The ground sub levels $m_s = \pm 1$ are degenerate in energy for symmetry reasons, but a local change in the symmetry can separate the degeneracy making them distinguishable. Note that the relative energies of the triplet states and singlet states are currently unknown.

electron spin, going from $m_s = \pm 1$ to $m_s = 0$. This anti-crossing is forbidden by symmetry considerations however is still experimentally observed.

Even though this anti-crossing behaviour is forbidden it turns out to be favourable as it is this anti crossing transition that provides the NV^- centre its ability for strong optical spin polarisation. This property is the crucial element for using the NV^- centre as a spin quantum bit or as a probe for high-resolution magnetometry.

The polarisation mechanism is explained in figure 3.5. Initially the system is unpolarised and the electrons fill the $m_s = 0$ and $m_s = \pm 1$ levels following the Boltzmann distribution (a). By optically pumping the transition with a excitation laser (e.g 514nm or 532nm), the population is excited from the ground state $m_s = 0$ and $m_s = \pm 1$ to the excited state (b). This excitation action is spin conserving so the population maintains its spin state at $m_s = 0$ or $m_s = \pm 1$. The system relaxes, mainly radiatively through the spin conserving 637nm optically active transition, however some part of the excited $m_s = \pm 1$ population relaxes through the metastable states and into the ground state $m_s = 0$ via a spin anti-crossing (c).



Figure 3.5: Schematic representation of the NV polarisation mechanism. (a) Initially the NV electron population is evenly distributed between the ground spin sublevels. (b) A ~532nm laser drives the optical transition and the NV population is excited into the excited states maintaining spin conservation $m_s = 0$ and $m_s = 1$, denoted as $|0\rangle$ and $|\pm1\rangle$ respectively. (c) The system relaxes, mainly radiatively by emitting photons at the 637nm zero phonon line. Some of the excited state with $m_s = \pm 1$ relaxes non-radiatively, into the singlet metastable states. They then continue to decay to the ground state in a non-spin conserving manner. (d) At the following cycle, the ~532nm laser again drives the optical transition conserving spin towards the excited state. (e) As in step (c) part of the population decays non-radiatively through the singlet metastable states to the $m_s = 0$ ground state. After a few optical cycles the NV spin population is therefore initialised into the $m_s = 0$ ground spin state. [66]

Note that the metastable states have been simplified to a single metastable state as the fine structure is not of direct concern in this case. After a few cycles the population of the NV⁻ ends up being polarised into the $m_s = 0$ ground state sublevel (e). This polarisation of the spin polarisation to $m_s = 0$ can be achieved for any initial spin population density. In this way the initial state of the system can be easily and reliably set or initialised (polarised) into a specific spin state necessary for further manipulation.

In addition the spin state of the NV⁻ transition can be optically read through measurements of the fluorescence of the ZPL. When there is a population in the $m_s = \pm 1$, a portion of the population relaxes through the non-radiative singlet states. The non-radiative relaxation mechanism causes a detectable drop (~ 30%) in the 637nm fluorescence of the NV⁻ centre compared to a state in which the electron always oscillates in an $m_s = 0$ sub-state.

Further manipulation of the ground-state spin can be achieved by applying a resonant microwave field at 2.88Ghz. Averaging in time the electron spin state population nutates between the $m_s = 0$ and the $m_s = \pm 1$ ground spin states due to the coherent interaction of the spin with the microwave field [51]. For each single NV⁻ the spin nutates between the $m_s = 0$ and $m_s = \pm 1$ spin state at frequency defined as the Rabi frequency. This nutation can be read out optically through the fluorescence intensity drop when the spin state is $m_s = \pm 1$. A new initialisation state can now be generated by applying a single pulse of a certain length known as a π -pulse. This takes the initialisation state of the system from an $m_s = 0$ spin state to a $m_s = \pm 1$ spin state.

In figure 3.6 the periodic modulation of the spin state is clearly visible in the fluctuations



Figure 3.6: Nutations of the electron spin between ground state sublevels of a single NV defect. The spin state has been initialised into an $m_s = 0$ spin state. Subsequently, a resonant microwave pulse of variable duration was applied. The state was read out optically. 10^6 measurements were accumulated to obtain a smooth curve. The fit curve represents an exponentially decaying harmonic function. [51]

of the fluorescence signal. These oscillations are called Rabi oscillations and correspond to the coherent oscillation of the electric spin between the $m_s = 0$ and $m_s = 1$ sublevels. In this experiment the $m_s = 1$ and $m_s = -1$ are separated due to strain in the diamond lattice and the microwave field is specifically tuned to the $m_s = 1$ state and hence nutation occurs only between the $m_s = 0$ and $m_s = 1$ sublevels. Upon increasing the pulse length, the fluorescence decreases, reaching the highest population inversion at a pulse duration of 160ns which corresponds to the π pulse.

In this work by completely characterising the optical forces due to a population of electrons being cycled in the 1042nm metastable transition we expect to provide new insights into the structure and behaviour of this state which is key to the use of NV centres as a quantum bit.

4

Experiment

In this chapter I present the experimental apparatus, procedures and technological solutions that I have developed in order to tackle various challenges in an attempt to build a platform with the potential to analyse the optical forces on optically trapped nanodiamonds containing many NV centres. Specifically I focus on developing a custom built optical trapping system and developing an imaging system to comprehensively monitor the motion of the particles within the trap. By carefully addressing the environmental noise in the apparatus I aim to minimise the uncertainty in measuring the optical forces on the trapped particles. Thereby providing a platform with the ability to accurately characterise the forces on atomic defects within trapped nanoparticles. Furthermore, I use this platform to investigate the classical and quantum forces on optically trapped nanodiamonds containing many NV centres at the 1042nm transition.



4.1 Optical Trapping Setup

Figure 4.1: Schematic representation of the optical trapping setup. A 1064nm trapping laser is incident on a half wave plate and polariser to control the power whilst maintaining horizontal polarisation. The laser beam profile is then cleaned up using a spatial filter (a) before reflecting off the polarising beam splitter, through the microscope objective onto the sample. The back reflected light from the sample is recollected by the microscope objective alongside some of the light being emitted by the goose neck lamp. The light recollected by the microscope objective is then directed through the second spatial filter (b) and focused onto the CCD camera for real time imaging. By switching the flippable mirror the light is filtered (Long pass filter cuts out the un-collimated goose neck lamp light) and imaged using the balanced detector setup.

4.1.1 Optical sources

A schematic of the custom built optical trapping setup is shown in figure 4.1. This setup is designed to optically trap individual particles and to monitor their motion. The trapping laser of the optical trap is a continuous wave 1064nm Neodynium:YAG laser (Ventus 1064, Laser Quantum). Continuous wave 1064nm Neodynium:YAG lasers are commonly used for optical trapping for a variety of reasons. Firstly optical traps are used extensively for biological applications and 1064nm is within the biological transparency window. Secondly

lasers of this type are commercially available at inexpensive costs with high power, high stability and excellent beam profiles. The specifications of the trapping laser are shown in figure 4.2.

SPECIFICATION	
Power	50mW - 5W
Navelength	1064nm
Beam Diameter	2.4mm ± 0.2mm
Spatial Mode	TEM00
Bandwidth	80GHz
)ivergence	<0.6mrad
A Squared	<1.5
ower Stability	<0.2% rms
loise	<0.2% rms
oise Bandwidth	1Hz - 100MHz
ointing Stability	<5µrad/°C
olarisation Ratio	100:1
olarisation Direction	Vertical
oherence Length	4mm
eam Angle	1mrad
perating Temp	10 - 40°C
Veight	1.3kg
mbilical Length	1.5m
Varmup Time	10 minutes
afety Class	Class IV

Figure 4.2: Specifications of the 1064nm trapping laser.

The laser was firstly directed through a half wave plate and polariser. By rotating the half wave plate the laser source power can be adjusted with the output remaining horizontally polarised. The beam profile was then cleaned using a spatial filter which consists of focusing the beam by a 35mm lens (LA1027-C, Thorlabs) through a 25μ m pinhole (P25C, Thorlabs) and re-collimating the beam using another lens of the same focal length.

Three mirrors (PF10-03-P01, Thorlabs) redirect the laser beam to be incident on the pellicle beam splitter (BP145B1, Thorlabs) angled at 45° to reflect the beam vertically towards the microscope objective. Part of the trapping laser power is reflected into the back entrance aperture of the water immersion microscope objective (UPLSAPO 60XW, Olympus) which focuses onto the prepared particle solution.

4.1.2 Trapping chamber

Particle solutions were diluted and placed into a micro-chamber made from two microscope coverslips stuck together with a commercially available double sided tape that produces a 20μ L chamber (UTIC-13-24x24, Bioscience Tools). The particle chamber is placed on an x-y stage (2-TSB28M, Thorlabs) at the focus of the microscope objective which is mounted on a z-axis stage (SM1Z) so the focus can be moved vertically throughout the entire chamber. The main nanoparticles that were trapped throughout the experiments was a nanodiamond solution made by diluting a sample of nanodiamonds of an average size of 100 ± 30 nm containing an average of 3000 NV centres. New nanodiamond chambers were made for each set of experimental results as the nanodiamonds solution was sonicated for at least 30mins to disperse the individual nanodiamond crystals.

The laser light transmitting through the microscope objective can now apply forces on the particles and then trap them at the focus of the laser. Particles that are contained in the cone of light but are below the focus are pushed, due the scattering force, into the focus of the trap. On the other hand particles that are contained in the cone of light above the beam focus are pushed away from the focus towards the top surface of the chamber where they will drift away due to Brownian motion. There is a need to be careful about bringing particles down to the bottom surface, since if you place the focus below the coverslip the particle will be pushed up out of the trap and the scattering force will continue to push the particle away from the trap. However once the particle is placed in this location other particles are unlikely to enter the trap, as the cone of light above the laser focus will push these particles away from the trap. This effect is shown in figure 4.3.

The particles that are trapped and held at the centre of the focus will strongly scatter photons. This backscattered light from the particle is transmitted through the pellicle beam splitter onto the detection beam path.



Figure 4.3: Diagram of where the particles are pushed in the chamber due to the scattering force. The scattering force is always in the direction of propagation which in this case is from bottom to top of the chamber. In this way trapping a particle at the bottom of the trap helps to ensure that other particles do not jump into the trap.

4.1.3 Beam splitter



Figure 4.4: Schematic of the behaviour of the system for beam splitters of different reflectance. In this case the sample reflects 35% of the incident beam. Figure (a) shows the system characteristics of a 50% reflective beam splitter. Figure (b) shows the system characteristics of a 10% reflective beam splitter. When laser power is limited using a 50% beam splitter maximises the output signal strength of the system. If there is sufficient of laser power to spare, using a less reflective beam splitter (b) gives us a much higher signal power for the same amount of power incident on the sample.

The choice of beam splitter is highly dependent on the power of the laser being used. If there is more than enough power, a beam splitter with low reflectivity is used to maximise the power on the detection path. If the power is a limiting factor then a higher reflectivity beam splitter is used to ensure that we have sufficient trapping power at the focus. In this setup we had a sufficient power and a low reflectivity beam splitter was used. An example of this effect is shown in figure 4.4

4.1.4 Microscope objective

Once the requirement for a laser has been satisfied the microscope objective is the most important element for an optical trap. The choice of objective directly determines the efficiency of the trap, being the stiffness versus the laser input power, which is a function of

both the NA and the transmittance of the objective. In addition to trap small particles the NA and beam profile must produce a sufficient trapping force to overcome the scattering force pushing the particle out of the trap. Typically in this case it takes a high NA objective (1.2-1.4 NA) to produce a high enough gradient profile to overcome the scattering force. Objectives of this type are all oil or water immersion objective, which also plays a role in the transmission of the laser. When there is a refractive index mismatch between the immersion fluid and the trapping medium the refractive index mismatch causes spherical aberrations in the tightly focused beam. These spherical aberrations negatively impact the gradient profile at the beams focus and hence, using a water immersion lens in an aqueous trapping medium can overcome the benefits of using an oil immersion lens with a higher NA. The vast majority of objective lenses are complex multi lens constructions designed specifically for imaging purposes using visible light, not for focusing high intensity infrared laser beams. Therefore the optical properties of different objectives in the infrared can vary widely. Unfortunately, manufacturers rarely supply the information regarding transmission characteristics in the infrared, and when they do it may be over estimated, since the objective transmission is measured using an integrating sphere and hence scattered, unfocused light will also be measured which in this case does not contribute to trapping. Of some interest are figures 4.5 and 4.6 which are two separate tables that investigated the transmission characteristics of a variety of microscope objectives.

When microscope objectives absorb the laser power the trapping system is negatively impacted in three ways. Firstly as a direct result the power in the trap focus is diminished compared to the available power at the back entrance of the objective. Secondly the limited available signal on the detector is also reduced as the light is back reflected through the objective. The last impact from an absorbing objective is that the maximum possible power that can be transmitted by the objective before damage occurs reduces as the absorbance increases. Unfortunately, microscope objectives do not give a damage threshold for maximum intensity through the objective, and they are too valuable to for us to experimentally determine this value. In summary the more the objective absorbs the more care that is needed with the input laser power, both in damaging the objective and receiving a sufficient trapping signal.

To determine the transmission of the objective two methods were utilised. The first method was to directly measure the light intensity using a power meter (S121C, Thorlabs) at the back entrance and after travelling through the objective. This method produced a consistent objective transmission for various input powers of $T_o = 33\%$. However, the objective is a water immersion lens and part of the laser power will be cut off due to the air gap between the objective and the power meter. To more accurately determine the transmission of the objective lens the reflected signal was investigated. The setup to measure the transmission is given in figure 4.7.

Manufacturer	Type designation	Part number	Magnification	Numerical aperture	Transmittance at 1064 nm (±2%)
Carl Zeiss, Inc.	Plan Neofluar	44 04 80	100×	1.30 oil	59%
Carl Zciss, Inc.	Plan Neofluar	44 04 66	63 ×	1.25 oil, iris	60%
Carl Zeiss, Inc.	Plan Apochromat	44 07 60	63 ×	1.40 oil	49%
Carl Zeiss, Inc.	Achrostigmat	44 02 55	40×	1.30 oil	49%
Nikon, Inc.	CF Fluor	85005	100×	1.30 oil	68%
Nikon, Inc.	CFN Plan Apochromat	85020	60×	1.40 oil	42%
Nikon, Inc.	CF Fluor	85004	40×	1.30 oil	74%

Figure 4.5: Transmission tables of commonly used microscope objectives for optical trapping experiments from Svoboda et.al. [4]

		Magnification/	agnification/	Transmission (±5%)			
Part No.	Manufacturer	Numerical aperture	Type designation	830 (nm)	850 (nm)	990 (nm)	1064 (nm)
461832	Zeiss	63/160/1.2 Water	Plan NeoFluar	66	65	64	64
506038	Leica	100/∞/1.4-0.7 Oil	Plan Apo	58	56	54	53
85020	Nikon	60/160/1.4 Oil	Plan Apo	54	51	17	40
93108	Nikon	60/∞/1.4 Oil	Plan Apo CFI	59	54	13	39
93110	Nikon	100/∞/1.4 Oil	Plan Apo CFI	50	47	35	32
93110IR	Nikon	100/∞/1.4 Oil	Plan Apo IR CFI	61	60	59	59
93144	Nikon	100/∞/1.3 Oil	Plan Fluor CFI	67	68	—	61

Figure 4.6: Transmission tables of commonly used microscope objectives for optical trapping experiments from Neuman et.al. [86]



Figure 4.7: Schematic representation to determine the transmission characteristics of a microscope objective. The initial laser power P_0 is measured just before the first interface of the beam splitter. This light is then incident on the beam splitter with reflectivity R_{BS} and transmission T_{BS} . The reflected light enters the objective with transmission T_0 and focused onto the mirror. The mirror reflects all of the light back through the microscope objective to transmit through the beam splitter and be detected by the power meter P. The transmission of the objective can then be calculated using equation (4.2).

The laser light is reflected off the beam splitter through the objective which is focused onto a mirror (PF10-03-P01, Thorlabs). The mirror is highly reflective at 1064nm and so reflects 100% of the focused laser light back through the microscope objective and onto the beam splitter. The final output power measured through the beamsplitter is then.

$$P_f = P_0 \cdot R_{BS} \cdot T_0 \cdot T_0 \cdot T_{BS}. \tag{4.1}$$

Rearranging for the transmission of the objective gives,

$$T_0 = \sqrt{\frac{P_f}{P_0 T_{BS} R_{BS}}}.$$
(4.2)

The input and output values were measured and the transmission of the objective was consistently measured to be $T_0 = 35\%$. This is consistent with the previous measurement, assuming 6% of the transmitted light will be lost in the water to air transition by total internal reflection. Unfortunately a value of $T_0 = 35\%$ is not ideal for a good efficiency and also reduces the maximum amount of power allowed in the objective. It was determined that the power in this objective should be limited to less than 200mW, since in this case 130mW is being absorbed by the objective which is getting close to where damage in the lens may start to occur, especially if the complex lens system focuses the beam throughout the multiple lens construction. Note that this is an arbitrarily chosen value and the microscope objective may be able to take more power than this, however higher powers increases the risk of damage.

4.1.5 Detection

Sensitive position detection is essential for a quantitative approach to optical trapping, since many physical properties of the trap such as force and trap stiffness can be determined by measuring the particle's position.

CCD camera

The first imaging system needed in the setup was a video based imaging system using a CCD camera (LU165M, Lumenera). The CCD camera's primary role is to determine if there was a particle trapped in the focus of the optical trap. When a particle entered the focus of the trap some scattered photons from the particle are collected by the microscope objective forming a diffraction limited Fourier image at the back entrance of the microscope objective. Using this re-collimated scattered light we could reconstruct a diffraction limited image on the CCD camera using Fourier optics as shown in figure 4.8. This allowed us to observe when a particle was trapped, which is necessary as there was no active mechanism to put the particle into the trap. Therefore I had to wait for tens of seconds to tens of minutes before a single particle would enter the trap focus through the Brownian forces in the fluid. One drawback of this method is that the longer you would like to characterise a single particle, the more dilute the sample needs to be to prevent the trapping of extra particles. However the more dilute the sample is the longer I would have to wait for an initial particle to enter the trap.



Figure 4.8: Schematic representation of the detection beam path showing the conjugate image (IM') and Fourier (FP') planes.

To increase the time to hold a single particle without other particles jumping into the trap it was essential to develop an approach to image a wide field view inside the solution, so that I could seek out individual particles in a very dilute sample. This was achieved through the use of a goose neck lamp (OSLI-EC Fibre Illiminator, Thorlabs) placed to illuminate the sample from above. The visible light shone down through the sample and was collected by the microscope objective lens and propagated through the detection path. Using the exact same Fourier optics used for the trapping laser the incoherent visible light provided a wide field image of the particles in the chamber. Diffraction limited profiles of the particles can be observed on the CCD camera, and odd shaped aggregates of particles can be easily identified and avoided. Particles of interest are then easily tracked and positioned at the focus of the trap by adjusting the x-y controls on the sample stage and z-control on the microscope objective mount. Once a particle is trapped at the centre of the trap the particle can be observed on the CCD camera as a high intensity, quickly oscillating diffraction limited spot. A snapshot of this image is seen in figure 4.9.

By digital processing the signal acquired from the camera, and knowing the size subtended by a single pixel, the position of the trapped particle can be determined with sub pixel



Figure 4.9: Images of the nanoparticles observed on the CCD camera. (a) The trapping laser is incident on the nanoparticle and is trapped at the focus. The bright spot at the centre of the image is a example of the quickly oscillating diffraction limited spot of the backreflected light of the nanoparticle. The goose neck lamp is also on giving a wide field image of the chamber. (b) The trapping laser is not on and two nanoparticles can bee seen drifting around the trap. The two particles appear as as different sizes, however this is due to their separate depths in the chamber. The static dots that do not change between the images are caused by dust on the CCD camera surface.

accuracy, using several centroid finding algorithms [87–89]. Video tracking of trapped objects has been implemented in real time, [90, 91] however this technique is limited by the frame rate of the camera (25Hz) and hence trapping frequencies near or above the frame rate of the CCD cannot be measured. To obtain a high frequency position measurement a quadrant photodiode (QPD) or high bandwidth balanced detector is needed.

Balanced detector

A balanced detector is a device that precisely measures a small difference in the intensity of two beams. A balanced detector consists of two photodetectors that measures the power of two incident beams and internal circuitry produces a voltage which is proportional to the difference in their intensities. The intuitive approach to how a balanced measures the position of a particle is shown in figure 4.10



Figure 4.10: Schematic showing how the position of the nanodiamond affects the Balanced detector signal. When the particle is displaced from the beam axis the beam power preferentially favours one detector and the difference signal shows a non zero reading.

When the particle is in the centre of the trap, the re-collimated back reflected signal is symmetric and centred on the centre of the back aperture of the objective. In this case a D-mirror (BBD1-E03, Thorlabs) splits the beam exactly in half so that the differential signal is zero. When the particle oscillates to one side of the trap the scattered signal from the particle is no longer symmetric on the back aperture of the microscope objective. In this case there is more intensity on one half of the beam relative to the other, and the Dmirror splits the intensity unevenly resulting in a voltage signal on the balanced detector. This signal increases the further the particle is moved from the centre of the trap. If the particle is moved to the other side of the trap a negative voltage is recorded on the balanced detector. In the literature it has been agreed that to obtain a better trapping signal the balanced detector D-mirror should be placed in a plane conjugate to the back focal plane of the objective [92–94]. In this case instead of relying directly on the intensity of the back scattered light, this method relies on the interference signal between the light from the scattering from particle and the light reflected from the cover slip. The benefits of this method are that the measured signal becomes insensitive to absolute bead position in the particle plane, and sensitive instead to the relative displacement of the bead from the laser beam axis [92]. However, in order to produce significant interference between the particle and the coverslip, the particle cannot be trapped in the centre chamber but close to an interface. Trapping in the centre of the trap is impractical for another reason which is that the signal from just the backscattered light from the particle results in low light levels at the balanced detector, ultimately limiting bandwidth and noise performance of the trapped signal. Therefore trapping close to a coverslip surface is desirable for an optimal signal on the balanced detector. An example of a voltage signal of a optically trapped nanodiamond



in an aqueous medium is shown in figure 4.11.

Figure 4.11: Position measurements of an optically trapped nanodiamond.

Quadrant photodiode

Initially a quadrant photo diode (PDQ80A, Thorlabs) was used, which, unlike the balanced detector provides normalised differential signals in both lateral dimensions as shown in figure 4.12. However the responsivity of the QPD was not high enough to provide a sufficient signal and determining the trapping frequency was not possible.



Figure 4.12: Schematic of a QPD detector array. with the corresponding formula for calculating the position of a particle within the focus of the trap. The trapping laser beam imparts a different power signal on each quadrant of the QPD. By using the equations shown the position of the particle at the centre of the optical trap can be calculated.

It is also noteworthy that in this experimental setup as shown in figure 4.8, the balanced

detector plane is not exactly conjugate with the back focal plane of the objective, but conjugate with the plane 1f from the back focal plane. This was due to a lack of space in the back reflected beam path. However since our microscope objective is an infinity corrected lens the back focal plane transitions to the image plane only at ∞ . In this case we assume that that the beam profile at the back entrance of the microscope objective is only barely different from the true back focal plane. This assumption is clearly valid as we receive a strong position signal as shown in figure 4.11.

4.1.6 Trapping frequency

The trapping forces acting upon a diamond sitting in the focus of an optical trap, can be approximated to a simple harmonic oscillator. The energy J, stored in the spring is equal to half the spring constant, κ , times the variance in motion,

$$J = \frac{1}{2}\kappa \langle x^2 \rangle. \tag{4.3}$$

By the equipartition theorem, the energy in the Brownian motion of the trapped diamond is equal to $\frac{1}{2}k_BT$, by setting these two energies equal and solving for the stiffness yields

$$\kappa = \frac{\langle x^2 \rangle}{k_B T}.\tag{4.4}$$

Calculation of the variance in the position is straightforward, however it requires an accurately calibrated position detector. A more useful method of measuring the trapping stiffness involves measuring the frequency spectrum of the Brownian noise exhibited by the diamond. In a fluid the mass of the diamond is so small that inertial forces are much weaker than those of hydrodynamic drag. In this case, the motion of the diamond is that of a massless, damped oscillator driven by Brownian motion.

$$\beta \dot{x}(t) + \kappa x(t) = F(t), \qquad (4.5)$$

where, $\beta = 6\pi\nu a$ is the drag coefficient of the diamond, ν is the viscosity of the surrounding fluid, a is the radius of the diamond and F(t) is the force arising from Brownian noise. The frequency spectrum of the Brownian noise source, $\tilde{F}(f)$, has zero mean and is essentially constant in amplitude,

$$|\tilde{F}(f)|^2 = 4\beta k_B T. \tag{4.6}$$

The Fourier transform of equation (4.5) is

$$2\pi\beta\left(\frac{\kappa}{2\pi\beta} - if\right)\tilde{x}(f) = \tilde{F}(f).$$
(4.7)

The power spectrum is then given by

$$|\tilde{x}(f)|^2 = \frac{k_B T}{\pi^2 \beta \left[\left(\frac{\kappa}{2\pi\beta}\right)^2 + f^2 \right]}.$$
(4.8)

Equation (4.8) is that of a Lorentzian with a corner frequency $f_c = \kappa/2\pi\beta$. Therefore the stiffness of the optical trap is given by

$$\kappa = 2\pi\beta f_c. \tag{4.9}$$

Optical traps typically work around one micron above the coverslip where hydrodynamic drag is altered by the proximity to the surface. The viscous drag on a sphere of radius a in a fluid with viscosity ν whose centre is a distance h above a surface is [4],

$$\beta = \frac{6\pi\nu a}{1 - \frac{9}{16}\left(\frac{a}{h}\right) + \frac{1}{8}\left(\frac{a}{h}\right)^3 - \frac{45}{256}\left(\frac{a}{h}\right)^4 - \frac{1}{16}\left(\frac{a}{h}\right)^5},\tag{4.10}$$

where, corrected values of the drag coefficient at different heights from the coverslip is given in table 4.1.

The height of the bead above the coverslip surface can be found by monitoring the axial detection signal as the surface is moved into contact with the bead [93]. In our case we will be comparing the ratio of two trapping signals and hence the absolute trapping stiffness value is not a factor in this experiment.

Table 4.1: Drag on a sphere or radius a at position h from a planar surface. The drag on the sphere at $h = \infty$ is given by Stokes' Law, $\beta = 6\pi\nu a$.

(\mathbf{h}/\mathbf{a})	Drag relative to $h = \infty$
1.01	2.97
1.10	2.36
1.25	1.92
1.50	1.62
1.75	1.47
2	1.39
3	1.23
4	1.16
5	1.10
10	1.06
50	1.01
∞	1.00



Figure 4.13: Initial power spectrum density plot of a trapped nanoparticle in the optical trap. The narrow spikes in the signal are caused by mechanical vibrations in the experimental setup. The MatLab program, Tweerzercalib 2.0 was used to generate the plot and fit the Lorentzian curve to the data (Solid line).

The trapping frequency is evaluated from the position measurements using a MatLab package designed for precise calibration of optical tweezers, tweezercalib 2.0 [95]. Figure 4.13 is an example of a power spectrum density produced by the tweezercalib 2.0 Matlab program. The program also fits a Lorentzian profile to the data to produce a value of the corner frequency of the trapped nanodiamond which gives you a value of the trapping stiffness through equation (4.9). The spikes in the data are undesired frequency resonances that are caused by various environmental factors. To produce an accurate corner frequency with a minimal error I had to address these resonant peaks caused by the environment.

4.1.7 Environmental Isolation

Environmental isolation is essential to provide the greatest possible sensitivity for measuring the corner frequency of an optically trapped particle. Environmental factors that influence the optical trapping measurements include, temperature variations, acoustic noise, mechanical vibrations and air convection.

Thermal fluctuations lead to slow, large scale drifts in the optical trapping signal. The effects of temperature variations can be explicitly observed to change the thermal kinetic energy term $(k_B T)$ in the power spectrum equation (4.8). The temperature fluctuations also implicitly alter the drag coefficient of the particle in the aqueous medium, $\beta = 6\pi\nu a$ where $\nu(T)$ if a function of T.

Mechanical vibrations arise from many devices, various cooling fans around the lab all produce strong mechanical vibrations. Similarly compressors and pumps can couple mechanical vibrations, even students walking through the corridors outside of the lab can induce mechanical vibrations strong enough to be detected. These mechanical vibration alter the beam path through the system ultimately leading to beam pointing issues, directly altering the power observed by each photodiode of the balanced detector.

Acoustic noise can shake many of the components of the experiment, leading to induce mechanical vibrations. The optics that couple the laser into the objective, the microscope objective, the sample and all the detection optics are vulnerable to vibrations caused by acoustic noise.

Air currents through the lab can also induce low frequency mechanical vibrations on mounted optics as well as various optical perturbations. For example beam deflections from gradients in refractive index produced by density fluctuations in the convected air can cause intensity variations across the balanced detector signal. Another factor that plays a role in the detection signal is that the signal is highly sensitive to light scattering form airborne dust. This is especially important in optical planes where the laser light is focused, and hence a small dust particle can block significant portions if not the entire beam profile.

When analysing the trapping signals various environmental factors can be identified and measures taken to minimise their effects. For instance slow thermal drift can be neglected if the trapping measurement is a short interval, however for measurements over the length of minutes thermal fluctuations can significantly shift the corner frequency causing systematic fluctuations in corner frequency. To reduce mechanical vibrations the experiment has been built on top of a large floated optical table (Newport RS4000) that offers mechanical isolation from the ground. The optics are also placed as close to the optical bench as feasibly possible and mounted so that they are mechanically rigid, thereby reducing the mechanical resonances and vibrations. All power supplies and fans are placed on the isolated gantry above the optical table with cables hung loosely to reduce the chance of any coupling of mechanical vibrations. A significant vulnerability of the setup to mechanical vibrations arises where the beam path is reflected vertically away from the optical bench. In this case the optics cannot be brought as close as possible to the table and strong resonant mechanical vibrations can couple to these taller structure.

In my setup, the initial power spectrum signal of nanodiamonds trapped in distilled water showed the expected Lorentzian trend but with many sharp peaks as shown in figure 4.13. The sharp peaks in the data are consistent with vibrational resonances in the setup producing strong signals a particular frequencies. When this trace was taken a small fan on a 532nm diode laser situated on the optical table was still functioning. Even though this fan was not making any sound or noticeable vibrations it was stopped from rotating and the signal was remeasured. The resulting signal is shown in figure 4.14.



Figure 4.14: Power spectrum density plot of a trapped nanoparticle in the optical trap after a single fan was prevented from operating. Note that the mechanical resonant spike at 800Hz has been removed compared to the plot from figure 4.13. The MatLab program, Tweerzercalib 2.0 was used to generate the plot and fit the Lorentzian curve to the data (Solid line).

It was observed from the power spectrum density that the resonance at approximately 800Hz was eliminated, which provided a better fit and more accurate corner frequency. From this point all possible sources of mechanical vibrations were systematically addressed. I placed silicon mats under many of the components to reduce the possibility of induced mechanical vibrations caused by air convection and acoustic noise. Those that were mounted higher off the optical bench required even more care as these were much more vulnerable to these induced vibrations. The result of placing the silicon mats and securing many of the optical components dramatically reduced the various resonant vibrational peaks in the power spectrum density. The resulting power spectrum density is shown in figure 4.15.



Figure 4.15: Power spectrum density plot of a trapped nanoparticle in the optical trap after all the mechanical vibrations were eliminated. The MatLab program, Tweerzercalib 2.0 was used to generate the plot and fit the Lorentzian curve to the data (Solid line).



Figure 4.16: Power spectrum density plot of an empty optical trap. The mechanical vibrations can be observed however they are now much smaller than the signal obtained from a trapped particle. The MatLab program, Tweerzercalib 2.0 was used to generate the plot and still attempted to fit a Lorentzian curve to the data (Solid line).

The power spectrum density was also measured when there was no particle in the trap. The power spectrum density no longer resembled the Lorentzian shape expected, and the fit is obviously incorrect. Noise was still visible in this case and arose because the environmental vibrations in the system were now the only cause of a fluctuating signal on the balanced detector, as shown in figure 4.16. This figure also demonstrates that the noise in the system has not been completely eliminated, but instead that the signal from the trapped particle is sufficiently large to hide the much reduced noise arising from environmental factors. In addition is is clear that low frequency noise is also much stronger than the rest of the noise. Unfortunately the low frequency noise usually cannot be removed from the setup as even the optical bench does not isolate against noise less than 7Hz.

4.2 Quantum-optical Trapping of NV centres in diamond

The aim of this section is to apply atom trapping techniques to apply an added restoring force caused by the NV centres contained within the nanodiamonds. Alongside this experiment, another part of the group was attempting to observe quantum trapping on the 637nm transition of the NV centres in diamond on a customised confocal microscope. My experiment was to observe quantum trapping in the 1042nm singlet state of the NV centre using a custom built apparatus. The custom built apparatus offers increased flexibility and a dedicated platform for studying a large variety of optical defects. The choice of the 1042nm transition was made for a few reasons. Firstly, the strength of the dipole is much larger than for the 637nm transition due to the much shorter upper level lifetime. Additionally this trapping mechanism will also provide a new way of studying the 1042nm metastable state, the structure of which is still debated in the literature. Finally, the 1042nm state cannot be optically addressed and therefore held by quantum trapping, without activation by an excitation laser exciting the 637nm ZPL transition. In this way this experiment will exhibit true quantum transitions. The reason the transition is optically inactive without the excitation laser is because all the electron sit in the degenerate ground states and hence there is no electron population in the 1042nm metastable state as shown in figure 3.5.

Once a population of electrons can access the singlet state the trapping laser is theorised to cycle the electrons in this state causing a force on the particle under the potential in equation (2.43). In this case the closer the laser wavelength is to the singlet transition the more efficiently electrons will be cycled in this state. We believe that it is the effect of cycling electrons in this state that causes the reduction in fluorescence of optically levitated NV centres in nanodiamonds [14].

The quantum force on the particle is a product between the trapping potential of a trapped NV centre as well as the number of NV centres that are efficiently cycled in the 1042nm transition of the NV centre. The number of efficiency cycled NV centres is expected to increase as the wavelength of the trapping laser approaches the 1042nm optical transition, however we do not know how much this cycling efficiency will increase and what shape this effect will display. The cycling efficiency of the system will also be affected by the variation of the transition wavelength from NV to NV within a single nano-diamond. The amount of wavelength variation of this transition is not known. In comparison, the 637nm ZPL transition distribution (NV to NV variation) has a standard deviation of $\sim 2nm$. In figure 4.17 the effect of the quantum force from the 1042nm transition in an ensemble of NV centres is plotted for three different populations of electrons in the 1042nm state. As mentioned previously the number of NV centres expected to be efficiently cycled and therefore applying a quantum force is unknown.



Figure 4.17: Plot of the ratio of the total force normalised by the classical force as a function of wavelength for the quantum optical trapping of various populations of electrons cycling in the 1042nm state. The transition is assumed to have a 1ns lifetime and a standard deviation between centres of 2nm, which is a value similar to the known broadening of the 637nm NV transition in nanodiamonds. A value of 24mW of power at the focal spot focused through a 1.2NA objective was also used, these parameters are important but not critical as the ratio changes only slightly with the incident power far from saturation of the quantum force. The dashed lines indicate wavelengths where the quantum force is the same magnitude as the classical force.

Initially, I planned on using a tunable femtosecond pulsed Ti:Saph laser to address wavelengths close to the 1042nm optical transition, however the light had to be fibred into the optical setup. The combination of the femtosecond pulses and the optical fibre created many problems that prevented the use of this laser. The major issues that prevented the use of the Ti:Saph was that only low powers could be coupled out of the fibre and that the spectrum of the light centred at a particular infrared wavelength was greater than 20nm. I decided to change tactics and use a tunable diode, yet, in this case there were delays in building the housing of the laser in order to cool the diode down to reach the 1042nm transition. The diode housing has only recently been completed and integrated into the setup and there has not been enough time to produce results. Due to this in the following results I will focus primarily on the preliminary results from the 1064nm trapping laser.



4.2.1 Experimental setup for addressing 1042nm transition

Figure 4.18: Schematic representation of the optical trapping setup for addressing the 1042nm transition. The 1064nm trapping laser and Diode probe laser are directed into a polarising beam splitter and focused into a single mode optical fibre. The output of the optical fibre is reflected from a dichroic mirror before reflecting off the beam splitter, through the microscope objective onto the sample. The 532nm excitation laser is fibred into the setup to clean the beam profile. The output of the 532nm single mode fibre is reflected off two mirrors through the dichroic mirror, so that the beam is collinear with the infrared beams. The back reflected light from the sample is recollected by the microscope objective alongside some of the light being emitted by the goose neck lamp. The light recollected by the microscope objective is then directed through the second spatial filter (b) and focused onto the CCD camera for real time imaging. By switching the flippable mirror the light is filtered (Long pass filter cuts out the un-collimated goose neck lamp light and 532nm excitation beam) and imaged using the balanced detector setup.

The first addition to the experimental setup was an excitation source to provide a population of electrons in the 1042nm singlet states. The NV^- excitation source was a continuous wave 200mW 532nm diode pumped solid-state laser.

This laser is chosen due to its low cost and high power. It is essential that the excitation

wavelength is within 450–575nm [96] so that the NV⁻ ZPL is excited and that the NV⁰ state is suppressed. The high power is needed so that it is easy to obtain enough power at the focus so that we can saturate this transition if desired. The laser is firstly directed through an objective lens (A375TM-A, Thorlabs) mounted on a z-stage to focus the beam profile onto on a xy-mounted (ST1XY-S, Thorlabs) single mode fibre (P1-488PM-FC-2, Thorlabs). The beam coming out of the fibre is collimated by an objective lens (LA1340-A-ML, Thorlabs) and then reflected by two mirrors (PF10-03-P01, Thorlabs) through a dichroic mirror (DMSP805, Thorlabs). The two mirrors were used to finely adjust the direction of the beam so that it was as close as collinear with the infrared beam path as possible.

Another important modification revolves around the fact that the system was designed and built to characterise the trapping behaviour of the nanodiamonds containing NV centres at the optically active infrared 1042nm transition. Hence we needed to have another trapping laser that spans the wavelengths around this region. Unfortunately in this specific region there is a lack of commercially available laser diodes to easily span this transition. We thus purchased the closest diode to the transition, which happened to be a Thorlabs 1060nm laser diode (L1060P200J, Thorlabs). This diode was stated to have a room temperature wavelength from 1040nm to 1080nm. The diode's actual room temperature was found to be 1061.9nm with a temperature dependence of 0.46nm/°C. From this we realised that at the lowest bound of the diodes working range (-20°C) the wavelength will be reduced to \sim 1041nm. Using this diode under a thermally controlled environment we can at least span the red half of the optically active infrared 1042nm transition. The diode was encapsulated in a custom built water cooled and water tight casing, to ensure that the diode could be cooled without risking water condensation. The casing contains all the electronics as well as the collimating objective lens (C230TMD-B, Thorlabs) so that the output of the casing is the desired collimated beam at the chosen wavelength. It was essential to have the beams of both this probe laser and the trapping laser completely collinear to ensure that the focuses are completely superimposed at the centre of the optical trap. The only possible way to ensure this outcome is to couple both lasers into the same optical fibre.

To do this the 1064nm laser was modified so that after it has been through the half wave plate and polariser, the two mirrors (PF10-03-P01, Thorlabs) redirect the laser beam to be incident on a polarisation beam splitter. The beam splitter reflects this beam due to its polarisation state onto an objective lens (C230TMD-B, Thorlabs) which focuses the beam into a single mode optical fibre (P3-980A-FC-2, Thorlabs). The two mirrors are carefully adjusted so that coupling of the fibre is maximised. The objective is held in a z-axis stage (ZM1Z, Thorlabs) in a cage system and optical alignment is optimised in conjunction with the two previous mirrors. The beam coming out of the fibre is collimated with an achromatic doublet objective lens (AC127-019-C-ML, Thorlabs) and is reflected off a dichroic mirror (DMSP805, Thorlabs) towards the pellicle beam splitter. An advantage of this system is that the fibre before the collimating objective lens can be replaced for a different fibre coupled laser source, for example if another optical transition needs to be addressed.

Secondly the collimated output of the tunable diode laser is set to be reflected off two mirrors that direct the beam onto the polarising beam splitter (BP145B2, Thorlabs). The polarisation was such so that the probe beam was transmitted through the polarising beam splitter allowing all of the light to be focused by the objective lens (C230TMD-B, Thorlabs)

to couple into the single mode fibre (P1-488PM-FC-2, Thorlabs). At this point the two laser modes of the 1064nm trapping laser and the diode probe laser are equivalent. Chromatic aberrations at the output are reduced by using an achromatic doublet collimating lens (AC127-019-C-ML, Thorlabs) at the output face of the fibre. The collinear beams are then incident on the dichroic mirror (DMSP805, Thorlabs) to combine with the excitation laser. The beam path through the rest of the system is then equivalent to the setup described in section 4.1.

4.3 Protocol

Developing a protocol for trapping and measuring the forces on the nanodiamonds within the optical trap is essential to produce accurate data that is consistent between measurement sets. The following steps were undertaken to ensure the highest accuracy and consistency of measured results.

Initially the trapping laser and excitation lasers were turned on and allowed to stabilise for at least one hour. If the lasers are not given sufficient time, the power output varies significantly affecting the trapping signal. In fact, as the 532nm laser is stabilising the output often looks as if it is blinking showing dramatic power fluctuations for significant intervals before diminishing to a steady state stable output.

Once the lasers had stabilised the beams were blocked and the objective was prepared with a water immersion and the sample placed above the objective, held up by the sample holder. The gooseneck lamp was turned on illuminating the sample which is then imaged on the Lumenera CCD camera. The sample was translated above the microscope objective lens to identify a single nanodiamond in the solution. When a nanodiaomnd had been identified it was translated to the centre of the optical trap. Once in position the beam block in front of the trapping laser was removed and the trapping laser light scattered from the nanodiamond is observed in focus on the CCD camera, confirming that the nanodiamond had been trapped.

The gooseneck lamp was turned off and removed from the mount to reduce both the temperature fluctuations in the sample and the induced mechanical vibrations on the optical apparatus. The focus of the laser was then slowly brought to either the bottom or the top of chamber. The benefits of using the bottom surface is that nanodiamonds drifting through the cone of trapping light are pushed away from the trap focus. However the nanodiamond can easily be lost by taking the focus of the trapping laser too low, into the coverslip which will then push the particle away from the focus and out of the trap. On the other hand when trapping on the top surface the nanodiamond will not be lost however other nanodiamonds that drift into the cone of light under the focus are pushed towards the focus of the trap where they will interfere with the trapping measurements.

Once the nanodiamond was trapped against the top surface or bottom surface the flip mirror was flipped down to allow the detection signal to be incident on the balanced detector. The trapping signal is observed on the oscilloscope (WaveSurfer 24MXs-B, LeCroy) and the position of the D-mirror is slightly adjusted until the mean of the position signal is zeroed. The signal can then be acquired for any period of time. In most cases the trapping signal was measured for a total of 160 seconds and split into 16 individual sections of 10 seconds

each. In this way the variation of trapping signal can be analysed as it varies with time. In addition the 160 second traces can be divided into various segments with different trapping lasers.

Using these 160s traces two procedures were developed to identify the different trapping force components on the nanodiamonds containing NV centres.

The first is when only two lasers are being used, the 1064nm trapping laser and the 532nm excitation laser which is used to populate the excited state. By splitting the 16 traces into two sets of eight we can gather statistics on two separate trapping conditions. The first 80s of the trace builds up statistics of the trapping frequency variation of just the trapping laser and second 80s builds up the statistics when the diamonds are being trapped by the 1064nm trapping laser as well as being excited by the 532nm excitation laser addressing the 637nm ZPL. This procedure can be visualised in figure 4.19a.

The second type of trapping protocol is used when three lasers are used. The 1064nm trapping laser and the 532nm excitation as well as a probe laser which has a tunable wavelength close to singlet transition state to investigate the trapping affects dependence on wavelength. In this case the trapping signal trace is split into four sections of 40s each. The first section is just trapping with the 1064nm trapping laser, the second adds the 532nm laser. The third trapping signal adds the third probing laser. For the last section the 532nm green laser is blocked leaving only the trapping laser and the probe laser. An easier way to visualise this procedure is shown in figure 4.19b.



Figure 4.19: Visual interpretation of the two protocols for the quantum-optical trapping experiments. The areas that are shaded indicate that the laser is incident on the nanodiamonds within the trap. (a) Protocol for when there is only the 1064nm trapping laser and the 532nm excitation laser. (b) Protocol for the experiments that include the tunable diode laser.

This procedure allows us to completely characterise the optical forces on the NV centres and nanodiamonds within the optical trap. The first section allows us to observe the trapping frequency from just classical trapping alone as the 637nm transition is not addressed. The second segment allows us to observe the trapping frequency variation when the NV centres are excited. In this case care needs to be taken when characterising these traces as the components of trapping can arise from both classical trapping from the green laser and atom trapping on the 1042nm state. The third section by adding the probe laser allows us to observe the additional atom trapping from the probe laser at wavelengths much closer to the atomic transition. In the last section when the NV centres are no longer excited, the atom trapping from the NV centres will no longer occur and only the classical components on the trapping frequency will be measured. By observing the trapping frequency change between traces one and two, and between traces three and four, the classical trapping components and atom trapping components can be characterised.

Unfortunately, optical trapping produces noisy signals and hence many traces needed to be taken to produce reliable statistics about the system. The trapping frequency is then evaluated from the position measurements files off the oscilloscope using a slightly modified version of the MatLab package tweezercalib 2.0.

4.4 Preliminary Results

Using the first procedure above a set of preliminary results was obtained trapping the nanodiamonds using both the classical and quantum forces. The first 80 seconds of the trace shows a corner frequency that only depends on the classical trapping stiffness arising from the 1064nm trapping laser,

$$f_c = \frac{\kappa_{cl}}{2\pi\beta}.\tag{4.11}$$

The power of the trapping laser was 100mW, which corresponds to a transmitted power of 35mW. In the second half of the trace 1mW of 532nm laser power excites the NV centres within the nanodiamond allowing access to the 1042nm transition. The corner frequency now depends on the classical trapping stiffness as well a quantum trapping stiffness arising from the action of the 1064nm laser on the 1042nm atomic transition

$$f_c = \frac{\kappa_{cl}' + \kappa_q}{2\pi\beta} \tag{4.12}$$

Where κ'_{cl} is the contribution due to both the 1064nm trapping laser and the 532nm excitation laser. These values are measured at ten second intervals and a mean of this value is taken. Now by taking a ratio of these values, and assuming that the 532nm excitation laser does not provide a trapping force we get,

$$\frac{\kappa_{cl}' + \kappa_q}{\kappa_{cl}} = 1 + \frac{\kappa_q}{\kappa_{cl}}.$$
(4.13)

In this way we obtain a value of the contribution due to the quantum trapping of the NV centres. This ratio also eliminates the high variability of the initial corner frequency. The standard deviation of the corner frequencies are then normalised by the classical corner frequency so that they can be directly compared to the ratio values. These values for the preliminary traces are shown in table 4.2.

Table 4.2: Preliminary data of quantum-optically trapped nanodiamonds containing NV centres. There was 35mW's of trapping laser power focused through the 1.2NA water objective and the NV centres were excited with 1mW of 532nm laser power. The nanodiaonds were trapped for 160s and were excited by the 532nm laser for the second 80s. The trapping ratio is the total corner frequency (classical and quantum) divided by the classical corner frequency. The normalised standard deviation is the standard deviation normalised by the classical corner frequency so that is can easily be related to the trapping ratio.

Trace	Classical Corner Frequency (Hz)	Total Corner Frequency (Hz)	Standard Deviation (Hz)	Trapping Ratio	Normalised Standard Deviation
1	26	27	3	1.025	0.1
2	47	51	6	1.071	0.1
3	45	45	2	0.985	0.04
4	47	51	2	1.091	0.05
5	82	88	3	1.076	0.04
6	96	102	5	1.059	0.05
7	14	14	1	0.985	0.07
8	40	38	2	0.954	0.05
9	39	39	2	1.001	0.06
10	27	27	1	1.002	0.05
11	28	28	3	0.982	0.09

Although this is limited data it can be observed that a number of traces show a clear increase in the trapping stiffness when the 532nm light excites the transition. The 532nm laser power could cause a classical trapping force accounting for this increase only if the focal points of the two lasers were completely aligned. If the focuses of the 532nm laser and the 1064nm trapping laser are perfectly superimposed the 532nm laser would exhibit an addition classical trapping force with a consistent 6% increase in all of the trapping traces which does not appear to be the case in the preliminary data. It is unlikely that the 532nm laser and 1064nm trapping laser are perfectly aligned as the 532nm laser was only co-aligned with the trapping laser through manual adjustments to the mirror mounts reflecting the path through a dichroic mirror. Small errors in alignment of the lasers at the back entrance aperture also correspond to relatively large displacements in the focal plane due to the tiny spot sizes of the beams. Additionally, not only are these two beams aligned with each other by hand, chromatic aberrations within the microscope objective will also displace the two focal points axially within the sample. Now even if the two focal spots slightly overlap in the diamond sample the two trapping forces will actually compete with each other which can even cause a decrease in the trapping frequency. It is clear that these effects are independent of the trapped object. The classical effect of the 532nm laser, if any, would have shown consistently in the data. Due to these effects it can be considered valid to assume that the 532nm laser provides negligible classical forces in the optical trap. The impact of the 532nm laser on the classical trapping is closely related to the relative position of the 1064nm and 532nm focuses. On the other hand, the excitation of the 637nm transition only requires relatively

small intensities. So even though the 532nm laser is not influencing the classical forces in the trap it will still excite NV centres in the nanodiamond. The increase in the trapping signal can then be caused by the 1064nm laser producing quantum forces due to the 1042nm atomic transition of the NV centre. An example of how the corner frequency varies in a single trace of an optically trapped nanodiamond is shown in figure 4.20.



Figure 4.20: Plot of the corner frequency as the nanodiamond evolves in time. The 532nm excitation laser was incident on the trapped nanodiamond from 80s onwards as indicated by the shaded region. The red squares are the corner frequencies measured for each 10 second interval, the black line is an indication of the actual trapping stiffness. The slow increase in corner frequency when the NV centres are being excited is due to the poor cycling efficiency.

It is observed in figure 4.20 that the observable increase in corner frequency does not occur immediately after the 532nm laser is incident on the nanodiamonds but takes considerable time to build up. This is again consistent with the 532nm not causing classical trapping as this would occur instantaneously. The slow delay could be caused by the inefficient cycling of the 1042nm state by the 1064nm laser. The laser wavelength is far from the atomic transition and hence the effective scattering rate is small, limiting the efficiency of cycling in the 1042nm atomic transition. In addition the 532nm laser may not be efficiently exciting an electron population into the 1042nm state due to the focal misalignment discussed above. The electron in the NV centres in this case will only occasionally transition into the 1042nm metastable state at which point the quantum trapping can begin to occur. Moreover only the NV centres whose spin is aligned in the $m_s = \pm 1$ sub-state can decay into the metastable singlet states where quantum trapping can occur. A combination of these effect may play a role in the delayed increase in the observable increase in trapping frequency.



Figure 4.21: The values from the preliminary data that show an increase in corner frequency have been averaged and are plotted on the curves showing the ratio of the quantum force as a function of wavelength. The transition is assumed to have a 1ns lifetime centred at 1042nm. The trapping behaviour is plotted for various standard deviations of the central wavelength between centres. For comparison a standard deviation of 2nm is the variation observed in the 637nm NV transition in nanodiamonds. A value of 35mW of power at the focal spot focused through a 1.2NA objective was also used which is consistent with the experiment. Using these assumptions the standard deviation between centres does not strongly influences the expected number of NV centres as we are a long way from the optical transition. Hence we arrive at a plot with only one data point and only one fitting parameter, which is the number of number of NV centres were cycling in the 1042nm state. The theoretical fits indicate that 200 NV centres were cycling on the 1042nm state.

The traces that show a clear increase in corner frequency when the 532nm laser is turned on show trapping ratio's between 1.059–1.091 corresponding with a 6–9% increase in trapping stiffness due to the quantum trapping. Unfortunately we could not estimate how strong the effect is expected to be due to the unknown number of electrons cycling in the 1042nm state causing the quantum force. In figure 4.21 the four values that show a clear increase in corner frequency have been averaged and are plotted on the curves showing the ratio of the quantum force as a function of wavelength in order to deduce the population of electrons cycling in the 1042nm state. The reason we used only the four values that show a clear increase in trapping frequency is to give an indication of the highest population of electrons that are cycling in the 1042nm state.

By assuming that the transition lifetime is 1ns and the transition we are observing is strictly the 1042nm state, figure 4.21 suggests that to observe a 6–9% increase in trapping signal, 200 NV centres have to have their electron cycling in the 1042nm state upon which the quantum force is acting. This value is not an unreasonable value since the nanodiamond sample is specified to have $100 \pm 30nm$ nanodiamonds containing ~3000 NV centres. In this case only 7% of the NV centres are being cycled in the 1042nm transition.

Trapping data and signals are inherently noisy due to the random nature of the Brownian motion and the accurate fitting of the power spectrum density produces its own variations in deciding the parameters to produce the optimal Lorentzian curve matching the data. Huge variations in corner frequency are also observed due to the size variation the nanodiamonds. This is because the force on the nanodiamond is proportional to the cube of the radius $(F_{cl} \propto a^3)$. It was interesting to note, that from the preliminary data if we plot the strength of the quantum trapping as a function of the corner frequency as shown in figure 4.22, we notice an interesting correlation. The data seems to suggest that the strength of the quantum trapping is correlated with the classical corner frequency and hence size of the nanodiamond.



Figure 4.22: Plot of corner frequency as function of trapping ratio. The trapping ratio indicates the strength of the quantum trapping in comparison to the classical trapping strength. The corner frequency is a measure of the size of the particle. It is interesting to observe from this preliminary data that the size of the particle is correlated with the strength of the quantum trapping. The blue curve is the linear fit to the data.

To investigate the cause of this trend let us assume that we have two particles of radius R and 2R. We know that the classical force is proportional to the volume $F_{cl} \propto a^3$ and so,

$$F_{cl}(2R) = 8F_{cl}(R). (4.14)$$

Now the quantum force is only dependent on the effective number of NV centres cycled by the 1064nm trapping laser at the 1042nm atomic transition ($F_q \propto \langle NV \rangle_{1064}$). However if we assume that there is an even density of NV centres throughout the nanodiamond matrix the effective number of cycling NV centres scales with the volume of the sphere so that,

$$F_q(2R) = 8F_q(R).$$
 (4.15)
The total force of the particles is now,

$$F_T(2R) = 8F_{cl}(R) + 8F_q(R) = 8F_T(R), \qquad (4.16)$$

and the ratio of forces in both cases should be equal and hence no correlation on the size of the particle should exist under these assumptions.

One feasible explanation that could explain this phenomenon is due to various strain mechanisms near the surface of the nanodiamonds. It has been shown that strain effects on NV centres within 10nm from the surface of diamond can cause the ND's to cease fluorescence [97]. Our nanodiamonds are ~ 100 nm in size and hence a 10 nm shell around the nanodiamonds constitutes a significant volume of the nanodiamonds in which the 1042nm transition may be inaccessible due to strain effects. The stated value of 10nm is where surface strain effects start to be observed for NV centre fluorescence at 637nm. The effects of surface strain on the properties of the electronic structure of the 1042nm transition within the NV centre is completely unknown. Therefore in this case a value for an effective depth at which we can assume that the NV centres are inaccessible due to strain effects is also unknown, however it would be safe to assume that it would be of the same order of magnitude. In any case we rely on the 637nm transition to access the 1042nm metastable state and so if the 637nm transition is not optically active, this is potentially enough to prevent access to the 1042nm state. Figure 4.23 displays the fraction of effective NV centres that can be addressed for an arbitrary effective surface strain depth. Note that the number of effective NV centres is reduced by half when the nanodiamonds radius is five times larger than the effective surface strain depth. For our 100nm nanodiamonds, if we use the 10nm value taken from the observation of NV fluorescence, 50% of the NV centres within the nanodiamond will be inaccessible due to surface strain effects. From figure 4.23 we can also observe that a larger particle displays a larger fraction of accessible NV centres than smaller particles and hence will exhibit a larger relative quantum force, consistent with the results in figure 4.22.



Figure 4.23: Accessible NV centres as a function of particle radius normalised by strain depth. As strain depth is closer to the size of the particle more NV centres are caught in the area that is within the strain depth and hence are not accessible for quantum trapping. As a benchmark, if the radius of the particle is only 10 times larger than the strain depth, then $\sim 50\%$ of the NV centres will inaccessible.

5 Conclusion and Outlook

Since their discoveries classical and atom trapping have both shown remarkable applications in various fields of science. Classical trapping is used most predominately in biological sciences with the manipulation of living and non-living cells [4]. The development of atom trapping has provided many techniques for cooling atomic systems to investigate the weird and wonderful behaviour of quantum physics. Recently however classical trapping is being pursued for its potential applications in the field of quantum science. In order to do this many well established techniques are being borrowed from atom trapping and being applied to classical trapping systems in order to reach a mesoscopic quantum ground state. As of yet no one has developed an optical trapping setup that combines both classical trapping forces with atom trapping forces. Usually this would be impossible to do because these two forces need trapped particles with mutually exclusive properties. However by carefully investigating the possible atomic forces on an artificial atom within a nanodiamond I have developed an experiment that indeed combines classical trapping forces with atom trapping forces.

5.1 Conclusion

In this thesis I focused on developing a custom built optical trapping setup as a platform for assessing the viability of measuring the atom trapping forces on artificial atoms embedded within nanoparticles. Specifically I investigated the strength of the atom trapping forces acting on the 1042nm metastable transition of many NV centres contained within an optically trapped nanodiamond. This technique provides a foundation for future developments by producing stronger optical traps with increased control on individual nanodiamonds. Additionally, in a system of this kind more well established techniques can be borrowed from atom trapping due to the availability of a narrow accessible electronic transition. With these techniques it may be possible to push the temperature of these large particles down into their

quantum ground state.

In building the optical trapping setup many environmental factors were observed, strongly impacting the measurements of the trapped particle. Before meaningful data could be extracted, the impact of temperature variations, acoustic vibrations, mechanical vibrations and air convection needed to be identified. Extensive effort was undertaken to reduce all these sources of noises on the data to reduce the error in the measurements down to a few percent.

Once the error was minimised, protocols for consistent measurement of the particle motion was developed and implemented to characterise the strength of the atomic trapping forces on the 1042nm transition of the NV centres embedded within optically trapped nanodiamonds.

A set of preliminary data provided insight into the optical forces acting on both the NV centres and the nanodiamonds. The preliminary data indicates an observable increase in the trapping stiffness when the 532nm laser excited the NV centres within the nanodiamond. The preliminary data also hints towards two other possible phenomena. The increase due to the atom trapping forces does not appear to occur instantly but after a significant period of time (~ 10 s). Additionally the data appears to show a correlation between the size of the particle and the strength of the quantum trapping increase. If this is indeed the case then surface strain effects could be impacting the electronic structure of the NV centres near the surface rendering these centres inaccessible to the trapping laser.

The combined contributions of classical trapping and atom trapping provide extra control over the optically trapped nanodiamond. This potentially opens a whole new field of research. Further studies should be directed towards better understanding the trapping mechanisms, in order to better assess and develop the wide range of potential applications based on the exploitation of these extra forces.

5.2 Outlook

5.2.1 Characterising the 1042nm state of the NV centre

The relevance in this research resides in having investigated and established some of the fundamental properties of the quantum trapping forces on optical defects in optically trapped particles. What is still unclear however is the exact structure of how the atomic forces build-up within the trapped nanoparticles. In the near future further studies should be focused on completely characterising the atomic forces acting on the 1042nm state in NV centres. The observed atomic force's dependence on nanodiamond size was an unexpected result, however it may be feasible that surface strain in the lattice may render NV centres in this region inaccessible. The atom trapping force's dependence on the exact excitation power of the laser is also an important factor. An investigation into the excitation laser power will give insight into how it can populate the optical transition and the time scales on how fast the electron population builds up in the 1042nm state. Finally the dependence on wavelength should be investigated with the tunable diode laser which is now available. Theoretically, we have shown an exact potential in equation (2.43) that describes the force acting on each NV centre, however the exact force also depends on how many NV centers are efficiently cycled

in the 1042nm transition. The number of efficiently cycled electrons is expected to increase as the wavelength of the trapping laser approaches the 1042nm optical transition, however we do not know how much this cycling efficiency will increase and what shape this effect will show. By completely characterising the atomic forces within the trapped nanoparticle it should then be possible to decipher the structural properties of the 1042nm transition. Using the quantum trapping potential in equation (2.43) we will be able to determine the quantum dipole transition strength d, the linewidth Γ , the atomic transition frequency ω as well as the average variation in transition frequency from NV centre to NV centre.

5.2.2 Quantum-optical levitation

The ideal platform for investigating these values is to optically levitate the nanodiamonds. This platform is currently being built in parallel, however the main difficulty in this setup it to initially trap a single stable particle. By levitating these nanodiamonds the environmental noise will be reduced as well as eliminating the possible influence of the fitting program. Due to the lack of Brownian motion the optically levitated nanodiamonds will exhibit a clear sharp peak on the power spectrum density plot corresponding to the trapping frequency of the nanodiamond, which will have a much smaller error in measurement.

A possible application arising from characterising the atom trapping forces of NV centres at the 1042nm transition, is to measure the strength of various evanescent fields. The evanescent fields can be characterised if the wavelength of the field can excite the NV transition (450–575nm [96]). By scanning the nanoparticle through the evanescent field, the trapping stiffness of the particle will increase alongside the intensity of the evanescent field corresponding to forces due to atom trapping.

5.2.3 Quantum-optical trapping on other optical defects

The current experimental apparatus was designed so that lasers sources can easily be substituted through the use of optical fibres. In this way the setup is ideal for further investigation of other optical transitions and defects. The optical defect for maximum atomic trapping forces is one with an electronic transition with a short upper lifetime and hence has an extremely well defined optical transition. Optical defects occur in many crystals however diamond itself has been found to contain over 500 optically active defects over the range of 170nm to 20μ m [68] and alone provides a rich source of available targets. One example is to quantum-optically trap the SiV centre in diamond at its ZPL at 737nm. It is expected that the strength of this transition is comparable to the strength of the 1042nm transition and is much easier to access, however this accessibility eliminates the degree of freedom of an external control that varies the strength of the quantum trapping caused by the 532nm excitation laser.

5.2.4 Quantum-optical trapping on other particles

Another avenue of research that can occur in the short term is to investigate the possibility of optically trapping particles other than those arising from defects in crystal structure. Possible targets include doped nanoparticles, fluorescent molecules and quantum dots. These

particles are used extensively in biological applications for fluorescent labelling of cells and organelles, in order to monitor processes within the cell such as function, regulation and position. However precise manipulation of single-molecules and simultaneous imaging has not yet been reached. The main limitation is that, to date, conventional biolabelling and current manipulating methods are two worlds apart, separated by different dimension-range limits. While biolabelling pushes towards smaller and smaller nanometre-sized particles to achieve superior resolutions, classical optical trapping relies on bigger (in biology, generally $1-0.1\mu m$) particles to work. By using a quantum-optical trap, much larger forces could be produced in order to sufficiently trap these fluorescent labels. However there are still foreseeable limitations in quantum-optically trapping these fluorescent targets. Whilst trapping doped nanoparticles and fluorescent molecules may be possible, these fluorophores tend to photobleach. The photobleaching rate will also be increased by the strength of the trapping laser. Provided the quantum trapping force is sufficiently large, then the laser intensity could be reduced, limiting the rate of photobleaching whilst still providing strong manipulation. Quantum dots however show another problem in that they display a high rate of blinking. Unfortunately if quantum dots were trapped using a quantum-optical trap and blinking were to occur, the optical transition will no longer be accessible to the laser and the quantum force would reduce to zero. If blinking does not influence the quantum trapping forces or if it can be eliminated, using quantum-optically trapped quantum dots could provide the solution to simultaneously imaging and manipulating molecules within biological cells.

5.2.5 Quantum-optical trapping towards new fluorescent labels

With the advent of the quantum-optical trap, I foresee the development of a new fluorescent label for manipulation and imaging within biological cells. Quantum nanosensors based on nanodiamonds containing NV centres could behave as the fluorescent sensors. The nanodiamond is extremely hard, inert and non-toxic, which are ideal features for its use in biological applications. Nanodiamonds can be readily synthesised with a large number of colour centres and with crystal sizes of less than 10nm. In addition the surface chemistry is readily modifiable, which allows for the targeting of specific proteins and molecules. The nanodiamond however is only a carrier for the NV centres embedded within. The NV centres possess bright and photostable fluorescence unlike many of the traditional biolabels. Whilst photo blinking has been observed in small nanodiamonds, it has been shown to be controllable [97]. In addition to using the NV centre as the optical sensor, the NV centres will act as the mediator for the quantum-optical trapping. Thus by using the increased forces due to the quantum forces we can hope to trap nanodiamonds a few times smaller than the current $\sim 0.1 \mu m$ size limit of classical trapping. The combination of quantum optical trapping with these new nanodiamond sensors will provide superior sensitivity and superior manipulation for studying the biological organelles and push towards single molecule measurements in cells.

5.2.6 Quantum-optical trapping towards more massive quantum states

Perhaps one of the biggest challenges of modern physics is figuring out how to realize and take advantage of strange quantum behaviours in progressively larger and more complex systems. Quantum-optical trapping introduces new trapping mechanisms that increase the trapping potential on the trapped particle. Additionally, well-established cooling techniques borrowed from atom trapping could be applied to the quantum-optical trap. These techniques include Doppler cooling, optical molasses, velocity selective coherent population trapping and Sisyphus cooling. By utilising this new quantum-optical trap and the well-established cooling techniques, it should be possible to reduce these relatively large trapped particles to their vibrational quantum ground state even at room temperature. By doing this a whole new range of novel applications will be realisable. One particularly poignant application of a macroscopic ground state is to consider the compatibility of the underlying concepts of space-time and quantum mechanics, through the investigation of quantum gravity. Contrasting theories on the route for quantum gravity, such as those from string theory and quantum loop gravity, all propose that there should exist a fundamental length scale, a lower bound to any output of a position measurement [98–102]. The common argument of this statement is based on the expectation that the high energies used in trying to resolve small distances will eventually significantly disturb the spacetime structure by their gravitational effects. One of the important impacts of a fundamental length scale is that there should be a modification to the well-known Heisenberg uncertainty relation. Pikovsky et al. [39], motivated by the principle of the deformed commutator relations proposed an elegant electromechanical scheme to probe quantum gravity and investigate the modification strength of such an uncertainty principle. The scheme relies on displacements of the massive mechanical oscillator in phase space. The strength of the deformations are expected to be measurable for these large trapped particles acting as massive mechanical oscillators in their quantum ground state.

The aforementioned applications of the quantum-optical trap only cover the surface of all the possible applications that will be used in the future. Traps of this kind have the true potential of a technology, whose core principles will be translated and adapted to many other fields of physics and life sciences.

List of Symbols

The following list is neither exhaustive nor exclusive, but may be helpful.

- $n_m \dots \dots$ Refractive index of the medium
- $n_p \ldots \ldots$ Refractive index of the particle
- m..... Ratio of the refractive indexes $m = n_p/n_m$
- P Power
- W_0 Beam waist
- \mathbf{r} Position vector
- \mathcal{E} Electric field vector
- E_0 Electric field strength
- ω Frequency of the electric field
- $\omega_0 \dots \dots$ Transition frequency
- $\hat{x}, \hat{y}, \hat{z}$ Unit vectors
- ϕ Phase of the electric field
- H..... Magnetic field vector
- Z_0 Intrinsic impedance of the medium
- S Poynting vector
- *I*..... Beam intensity
- $\tilde{x}, \tilde{y}, \tilde{z}$ Normalised spatial coordinates
- $\langle \hat{\mathbf{D}}_{\epsilon} \rangle$ Expectation value of the dipole moment
- α Polarisability
- α' Real part of the polarisability

- α'' Imaginary part of the polarisability
- σ Cross section for the radiation pressure of a particle
- U..... Trapping potential
- $\Gamma \ldots \ldots \ldots$ Transition linewidth
- d..... Dipole moment of a two level transition
- s..... Saturation parameter
- T_0 Transmission of the microscope objective
- κ Optical trap stiffness
- β Drag coefficient
- $f_c \dots \dots$ Corner frequency
- $h \dots Height$ from a surface

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