

Forces in the Interaction of Light with Matter

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Abstract: It is well established that the interaction of light with matter generally imparts mechanical forces and associated torques. Where laser light is involved, such effects can be deployed for the microscale and nanoscale manipulation of matter, giving rise to notable applications in wide areas of physics, chemistry, and the life sciences. The diversity of applications is enriched not only by the complexity of the constitution of matter, but also by the structure of the laser light itself, according to its polarisation, amplitude, and phase. This article reviews in comprehensive terms the origins of laser-based optical forces and their manifestations in a variety of current contexts and applications.

Keywords: radiation pressure; optical force; laser manipulation; atomic gases; optical tweezers

1. Introduction

It has long been known that light imparts a pressure when it strikes an object, a phenomenon mostly evident at high levels of illumination. The mechanism for such effects first became comprehensible with the formulation of Maxwell's theory of light [1], which showed that in addition to carrying energy, a light wave carries linear momentum. Later experimental work by Lebedev [2] and then Nichols and Hull [3] confirmed the predictions of Maxwell's theory. Further developments culminated in work by Poynting [4] on the concept of optical momentum density. For light propagating in a vacuum, the formula for momentum density is simply W/c , which is the energy density W divided by the speed of light c . In the case of light travelling in a medium, however, from subsequent theory, it appeared that a corresponding expression for the momentum density could be cast in one of two forms separately advocated by Abraham and by Minkowski [5,6]. Abraham predicted a decrease in linear momentum as compared to the value on propagation in a vacuum, while Minkowski's formulation predicted a correspondingly proportional increase. The two corresponding formulae for momentum density, ρ^A and ρ^M , for a plane wave of frequency ω travelling along \hat{z} in a homogeneous, isotropic medium of refractive index n , are as follows:

$$\rho^M = \frac{n^2}{c^2} \mathbf{E} \times \mathbf{H} = n^2 \rho^A = \frac{n}{c} W \hat{z}. \quad (1)$$

In quantum terms, for a single photon $W = \hbar\omega$, we then have explicitly

$$\rho^M = n\hbar k \hat{z}; \quad \rho^A = \frac{\hbar k}{n} \hat{z}, \quad (2)$$

where ω/c .

All of these early studies predated the arrival of a quantum theory of light. Some of the puzzles that then led to a fully fledged theory based on Einstein's photon concept are nicely illustrated in subsequent work by Wheeler and Feynman in 1946 [7]. However, following the full establishment of quantized radiation theory, a substantial body of research resumed the debate on this Abraham–Minkowski controversy [8–15], and it was driven by



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laser experimentation. Nonetheless, the outcomes of these studies could not categorically determine the correct form of the momentum density. More recent work again revived the controversy, seeking its resolution [16–21]. Barnett and Loudon argued that the two densities are appropriate in different contexts. The momentum of a photon in a dielectric medium is not directly measurable; only the total momentum of the material and light combined is detectable. Accordingly, the division of total momentum into component (optical and material) parts may be considered arbitrary, this division being performed to signify either a kinetic or canonical momentum [22,23]. The former has its usual kinematic sense, associated with distinct kinetic energy: the latter, defined by Lagrangian mechanics, additionally accommodates the electromagnetic influence of the medium.

In classical terms, experimentally observable optical forces arise in consequence of Newton's Second Law of Motion through changes in the linear momentum of a beam as it interacts with matter—for example, through absorption, reflection, or refraction. Similarly, optically induced torques arise in consequence of changes in angular momentum. Since the early days of laser science, research has increasingly explored how optical forces might control matter in sizes ranging from the microscale down to nanoparticles and single atoms [8,18,20,24–56]. Ashkin and co-workers [57] were the first to show how to trap and manipulate small particles using laser light [58–72]. The emergent technique known as 'optical tweezers' has become a powerful tool for the manipulation of nanomaterials including molecular and biological matter [73–86]. The development of structured light, including *optical vortices* (or *twisted light*) has extended the capacity to characterise, trap, and manipulate small particles, including atoms and molecules [56,74,87–93]. Nanoparticles can also be trapped using strong plasmonic electric field gradients [82,94,95].

In general, mechanical effects can be considered to deliver two types of optical forces, namely the *scattering* or *dissipative* force and the *gradient* force. Focussing primarily on the extensive field of nanoscale applications, the development of theory to describe the mechanisms leading to optical forces have mostly focused on small dielectric particles, considered as Rayleigh particles. The primary assumption is that these particles acquire a fluctuating induced dipole moment on being subjected to the electric field \mathbf{E} of incident light. The induced moment then couples to the field gradients, leading to a gradient force. The dissipative force, on the other hand, arises as linear momentum transfers from the incident beam of light to the particle—the rate of momentum transfer determining the force. For an electric dipole, a third kind of optical force exists as a non-conservative scattering force associated with the curl of the spin angular momentum density of the light field [84]. Such mechanisms are valid for most applications of optical nanomanipulation, but for particles of dimensions commensurate with the wavelength, the Lorentz–Mie theory becomes the uppermost formulation, as recently discussed [96].

Whereas optical tweezers essentially employ spatially structured light fields to produce field gradients, which are then used to trap and manipulate small particles, other more subtle effects reported in the literature include chiral forces, in which magnetic dipole and electric quadrupole forms of coupling play a role alongside the electric dipole [96–118]. Such forces usually arise with circular or elliptical polarisations, associated with their spin [97–118]. Using the chiral properties of vortex-structured light carrying orbital angular momentum greatly expands the scope for chirally sensitive applications [56,74,87–93,96,119,120], although experimental achievement remains highly challenging.

Radiation pressure due to laser light is also engaged for the cooling and trapping of atoms, playing a particularly prominent role in cold atom physics. Atom trapping using laser light was first realised and explicated on the basis of an optical dipole force [121] arising from the off-resonance interaction of the atoms with the beam. While the scattering force is responsible for atom cooling [122], it can also contribute to atom trapping—for example, when the scattering force acts together with a magnetic field [123] or with the optical dipole force [121,124]. In systems comprising more than one particle, in addition to the optical trapping forces that act on individual components, additional optically induced inter-particle

forces can arise, leading to pairwise ‘optical binding’ phenomena [98,100,107,125–132] (see Section 5).

2. Optical Forces—Fundamentals

2.1. Optical Forces on an Electric Dipole

Optical forces on matter primarily owe their origin to dipolar interactions with light. In the quantum formulation, each particle is characterised by an electric dipole operator $\mathbf{d} = e\mathbf{q}$, where e is charge, and \mathbf{q} is a displacement vector. The quantum operator representation provides for the description of both fluctuating and transition moments. The simplest formulation that directly embodies the salient features of light–matter interaction is a quantum electrodynamical Hamiltonian theory [35,93,133]. The system Hamiltonian is here represented as follows:

$$\mathcal{H} = \frac{|\mathbf{P}|^2}{2M} + \mathcal{H}_a + \mathcal{H}_{fields} + \mathcal{H}_{int}, \quad (3)$$

where \mathbf{P} is the linear momentum of a particle of mass M so that the first Hamiltonian term is the kinetic energy of the centre of mass. The rest of the terms are as follows: \mathcal{H}_{fields} is the electromagnetic field Hamiltonian field; \mathcal{H}_a is the Hamiltonian representing the internal energetics of the particle, and \mathcal{H}_{int} is the interaction Hamiltonian, which in the electric dipole approximation is expressible as:

$$\mathcal{H}_{int} = -\mathbf{d} \cdot \mathbf{E} + \frac{1}{2M} [\mathbf{P} \cdot (\mathbf{d} \times \mathbf{B}) + (\mathbf{d} \times \mathbf{B}) \cdot \mathbf{P}]. \quad (4)$$

The velocity of the centre of mass $\dot{\mathbf{R}}$ follows as a Heisenberg equation

$$\dot{\mathbf{R}} = \frac{i}{\hbar} [\mathcal{H}, \mathbf{R}] = \frac{1}{M} (\mathbf{P} + \mathbf{d} \times \mathbf{B}), \quad (5)$$

where a superscript dot denotes a time derivative. The mass–acceleration product for the centre of mass is then given by

$$M\ddot{\mathbf{R}} = \frac{i}{\hbar} [\mathcal{H}, (\mathbf{P} + \mathbf{d} \times \mathbf{B})] = \nabla(\mathbf{d} \cdot \mathbf{E}) + \frac{d}{dt}(\mathbf{d} \times \mathbf{B}). \quad (6)$$

This expression represents the optical force \mathbf{f}_d acting upon an electric dipole. Note that the spatial derivatives in ∇ are taken with respect to the centre of mass variable \mathbf{R} . The time derivative in Equation (6) contains two terms, and with the use of the Maxwell equation, we find

$$\mathbf{F} = (\mathbf{d} \cdot \nabla)\mathbf{E} + \dot{\mathbf{d}} \times \mathbf{B}. \quad (7)$$

As shown below, this result can be generalised to the optical force in the many-dipole case, forming a medium. Before we move to consider optical forces in media, we briefly comment on the two terms involved in the optical force on a single dipole, as given by Equation (7). The first term is the dominant contribution to the force on the dipole, while the second term signifies an additional force that arises from the interaction term in the Hamiltonian. The latter, called the Röntgen force, whose origin has been established within the dipole approximation [133], has been the subject of interest in a number of recent investigations [56,133]. Surprisingly, it produces a momentum-dependent interaction that leads to high-field-seeking atoms being repelled from intensity maxima in a travelling laser pulse.

2.2. Optical Forces in Dielectric Media

Given their mode of operation on particles in free space, it is not surprising that optical forces may also have an effect on condensed phase matter. They can, for example, lead to electric phenomena such as the photon drag effect, in which a current is induced

due to light forces acting on quasi-free electrons in semiconductors and other dielectric media [47,51]. Optical forces in some dielectric media can be regarded as arising from the response of a distribution of microscopic atomic electric dipoles, representing the source of polarisation \mathcal{P} , subject to electric and magnetic fields \mathbf{E} and \mathbf{B} . We can then associate the following optical force density with the effect on an individual dipole \mathbf{d} upon being subjected to electromagnetic fields:

$$\mathbf{f} = \delta(\mathbf{r} - \mathbf{R})(\mathbf{d} \cdot \nabla)\mathbf{E} + \dot{\mathbf{d}} \times \mathbf{B}\delta(\mathbf{r} - \mathbf{R}). \quad (8)$$

The individual polarisation field of a single dipole is $\mathbf{p} = \mathbf{d}\delta(\mathbf{r} - \mathbf{R})$. This leads to the generalisation of the Lorentz force acting on a homogeneously distributed ensemble of such dipoles forming a dielectric medium, in which case the force density acting on an individual dipole of the ensemble is

$$\mathbf{f}_d = (\mathbf{p} \cdot \nabla)\mathbf{E} + \dot{\mathbf{p}} \times \mathbf{B}. \quad (9)$$

The dielectric medium is modelled as a set of N elementary dipoles per unit volume such that, aside from Lorentz local field corrections, the medium polarisation density is $\mathcal{P} = N\mathbf{p}$; \mathbf{p} is proportional to the electric field acting on the dipole $\mathbf{p} = \alpha\mathbf{E}$, where α is the dipole polarisability. So, we now have for the medium polarisation

$$\mathcal{P} = N\alpha\mathbf{E} = \chi\mathbf{E}. \quad (10)$$

where we have defined the linear susceptibility $\chi = N\alpha$. The optical force in the dielectric medium is

$$\mathbf{F}_{dielectric} = (\mathcal{P} \cdot \nabla)\mathbf{E} + \dot{\mathcal{P}} \times \mathbf{B}. \quad (11)$$

The polarisability and its bulk counterpart susceptibility are, in general, complex. On substituting for \mathcal{P} , we have, assuming that $\alpha = \alpha'$ is real,

$$\mathbf{F}_{dielectric} = \frac{1}{2}\alpha'\nabla|\mathbf{E}(\mathbf{r},t)|^2 + \alpha'\frac{\partial}{\partial t}\mathbf{E}(\mathbf{r},t) \times \mathbf{B}(\mathbf{r},t). \quad (12)$$

In practice, we require the cycle average of this force, which we denote as $\langle \mathbf{F}_{dielectric} \rangle$ and which is given by

$$\langle \mathbf{F}_{dielectric} \rangle = \frac{1}{2}\alpha'\nabla|\mathbf{E}|^2, \quad (13)$$

where we have set the cycle average of the second term in Equation (12) to zero. On the other hand, if the polarisability is considered to be complex, as befits near-resonant radiation, then $\alpha = \alpha' + i\alpha''$, and the force expression in Equation (13) becomes

$$\langle \mathbf{F}_{dielectric} \rangle = \frac{1}{2}\Re\{[(\alpha' + i\alpha'')\mathbf{E} \cdot \nabla]\mathbf{E}^*\}. \quad (14)$$

This expression contains both gradient and dissipative forces.

2.3. Rival Optical Forces in Media

Barnett and Loudon [49] contrast the above Lorentz force density \mathbf{f}_d (which is based on considering the dielectric as an ensemble of individual dipoles) with another force density \mathbf{f}_q in which the polarisation is determined by the Lorentz force on individual charges [46,47,134]. Here, we have

$$\mathbf{f}_q = -(\nabla \cdot \mathcal{P})\mathbf{E} + \dot{\mathcal{P}} \times \mathbf{B}. \quad (15)$$

It is straightforward to show that Equation (15) reduces to the Lorentz force density on expressing the charge density ρ and current density \mathbf{J} in terms of the polarisation density

$$\rho = -\nabla \cdot \mathcal{P}; \quad \mathbf{J} = \dot{\mathcal{P}}. \quad (16)$$

The force densities \mathbf{f}_d and \mathbf{f}_q look rather different, and suggestions were made that they should lead to different predictions that could be tested experimentally to determine which is physically correct. Barnett and Loudon [49] demonstrated that the volume integrals of the densities, on substituting $\mathcal{P} = \mathbf{d}\delta(\mathbf{r} - \mathbf{R})$, as is appropriate for a single dipole, in fact lead to identical expressions for the total forces $\mathbf{F}_d = \mathbf{F}_q$ —proving that the two formulations furnish the same predictions in a wide majority of contexts.

2.4. Two Types of Optical Force

Returning to the case of atoms, the electric field of the laser radiation in general possesses both an amplitude function \mathcal{U} and a phase function, $e^{i\Phi}$, so we have

$$\mathbf{E} = \mathcal{U}e^{i\Phi}. \quad (17)$$

Substituting, and extracting the real parts only, we obtain the sum of the gradient force and the dissipative force

$$\langle \mathbf{F}_{die} \rangle = \langle \mathbf{F}_{gradient} \rangle + \langle \mathbf{F}_{dissip} \rangle, \quad (18)$$

where

$$\langle \mathbf{F}_{gradient} \rangle = \frac{1}{4}\alpha'\nabla|\mathbf{E}|^2; \quad \langle \mathbf{F}_{dissip} \rangle = \frac{1}{2}\alpha''|\mathbf{E}|^2\nabla\Phi. \quad (19)$$

These expressions are valid for point-like particles. (When, in Section 4, we consider optical tweezer forces on potentially much larger particles, we shall use a less restrictive formulation).

To date, we have assumed a constant (i.e., frequency-independent) polarizability. However, in the interaction of atoms with light—especially in the simplest case of a two-level neutral atom—the polarizability exhibits classic dispersion under near-resonance conditions [135]. The scattering force which then arises results from the atom experiencing repeated cycles of excitation and spontaneous emission, accompanied by centre of mass recoil. Such processes result in a random distribution of the atomic linear momentum [93,135–137].

3. Optical Forces in the Laser Cooling and Trapping of Atoms

The atom is a neutral particle which can primarily interact with resonant electromagnetic fields through three basic processes, namely: *stimulated photon absorption*, *stimulated photon emission*, and *spontaneous photon emission*. When the atom interacts with a laser beam, there are two particular issues that have to be taken into account. First, the laser provides a flux of photons whose levels of intensity can cause these elementary interactions to be repeated at phenomenal rates. Second, no laser light is entirely monochromatic; generally, its spectrum has a width determined by the laser medium, ancillary optics, and Fourier constraints imposed on any pulsing. One result of these considerations is that the laser radiation pressure force should be considered as comprising two different types of force. It is important to recognise limitations on the validity of assuming that changes in atomic motion arise from a mean radiation pressure force acting on the atom (due to transfer of momentum in a specific position). This assumption is valid only if one can construct atomic wave packets with sufficiently small spreads, $\Delta\mathbf{R}$ and $\Delta\mathbf{V}$, without any violation of Heisenberg relations. A sufficient condition for this requirement is $\hbar\Gamma \gg E_{rec}$, where $E_{rec} = \hbar^2k^2/2M$ is the recoil kinetic energy due to photon absorption or emission.

3.1. Scattering Force, Doppler Cooling and Optical Molasses

The scattering force (or *dissipative force*) is generated by the atom scattering input photons, repeated cycles giving rise to a huge number absorption-spontaneous emission processes—typically about 10^8 such cycles per second. As the resonant scattering removes photons from the input beam, conservation of the linear momentum in the beam–atom system results in a loss of momentum for the beam and a corresponding gain for the

atom in the direction of beam propagation. For example, at typical levels of laser intensity and vapour density, one might expect a sodium atom, excited on its $3^2S_{(1/2)} - 3^2P_{(3/2)}$ transition, to experience an acceleration $a = N\hbar k/M \approx 10^5 g$, where g is the acceleration due to gravity.

The general formula for the scattering force is:

$$\mathbf{F}_{sc} = \hbar\Gamma\Omega^2(\mathbf{R}) \frac{\nabla\Theta(\mathbf{R})/4}{\Delta^2(\mathbf{R}, \mathbf{V}) + \Omega^2(\mathbf{R})/2 + \Gamma^2/4} \quad (20)$$

where $\Omega(\mathbf{R})$ is the Rabi frequency, $\Delta(\mathbf{R}, \mathbf{V})$ is the detuning (which, in general, may depend both on the atom's position and velocity, due to a Doppler shift), and $\Theta(\mathbf{R})$ is the phase of the electric field of the input beam [93]. When the laser input has the form of a Gaussian beam, then $\nabla\Theta(\mathbf{R}) \approx \mathbf{k}$, and the scattering force is mainly along the beam propagation direction. However, if the laser beam is in the form of a vortex of topological charge (winding number) ℓ , then $\nabla\Theta(\mathbf{R}) \approx \mathbf{k} + \ell\hat{\phi}/\rho$; in this case, the scattering force is also responsible for the generation of a torque acting on the atom. Here, when the laser intensity is relatively strong, the scattering force saturates to the expression $\mathbf{F}_{sc,max} = \hbar\Gamma k\hat{\mathbf{k}}/2$, while the torque reduces to the expression $\mathbf{N}_{sc,max} = \hbar\Gamma\ell\hat{\mathbf{k}}/2$.

If the atom moves with a velocity \mathbf{V} and is irradiated by a laser beam of angular frequency ω_L , it experiences a Doppler-shifted frequency $\omega_L - \nabla\Theta \cdot \mathbf{V}$; irradiation by a Gaussian beam thus results in a Doppler-shifted frequency $\omega_L - \mathbf{k} \cdot \mathbf{V}$. However, in the case of irradiation by an optical vortex, the shifted frequency is approximately equal to $\omega_L - \mathbf{k} \cdot \mathbf{V} + \ell V_\phi/\rho$, where the last term represents an *azimuthal Doppler shift* [138].

When a moving atom is subject to two weak counter-propagating Gaussian beams, it experiences Doppler-shifted frequencies for the two beams approximately given by $\omega_L \mp \mathbf{k} \cdot \mathbf{V}$. This frequency imbalance is responsible for the appearing of a damping force that is always proportional and opposite to the atomic velocity. This Doppler cooling can result in the formation of an ostensibly viscous atomic system known as *optical molasses* in one, two, or three dimensions [121]—see Figure 1. In this case, the total scattering force on the atom is given by $\mathbf{F}_{sc} \approx -a_z V_z$, with the damping coefficient given by $a_z = 2\hbar k^2 s \frac{(-\Delta\Gamma)}{(\Delta^2 + \Gamma^2/4)}$, where $\Delta < 0$, and s is the so-called saturation parameter given by $s = \frac{\Omega^2/2}{(\Delta^2 + \Gamma^2/4)}$. By such means, optical molasses can cool atomic motion up to a kinetic temperature known as a *Doppler temperature* [122], which is given by

$$T_D = \frac{\hbar\Gamma}{2k_B}, \quad (21)$$

where k_B is the Boltzmann constant.

The mechanism for optical molasses has a very interesting counterpart in the case of an atom interacting with optical vortices. If we assume that the moving atom is irradiated by two co-propagating optical vortices with opposite winding numbers, then we may observe so-called *azimuthal molasses*. In this case, the atom experiences a damping force which cools it in the azimuthal direction, given by $\mathbf{F}_{sc} \approx -a_\phi V_\phi$, with the damping coefficient given by $a_\phi = 2\hbar(\ell/r)^2 s \frac{(-\Delta\Gamma)}{(\Delta^2 + \Gamma^2/4)}$, where s is again the saturation parameter. In the case of two counter-propagating twisted beams with opposite winding number, we can secure a combination of axial and azimuthal optical molasses.

The dissipative force is in practice a coarse-grained average, since it is the result of numerous absorption–spontaneous emission cycles, and is consequently accompanied by fluctuations related to both the emission and the absorption processes responsible for a diffusion of the atomic momentum.

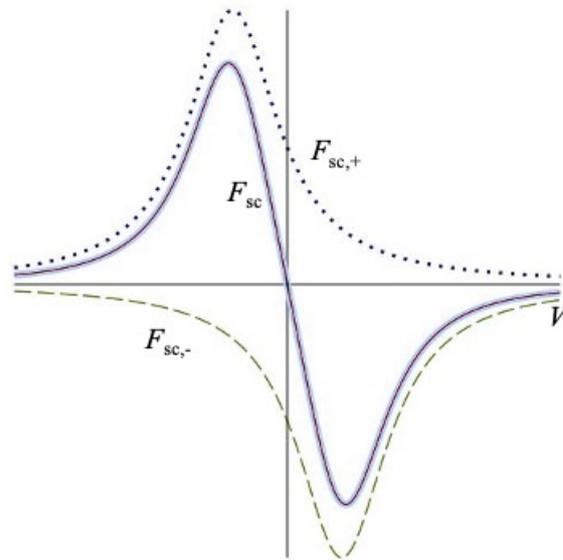


Figure 1. For a two-level atom moving with velocity V in the standing wave of two counter-propagating waves, the frequencies of the counter-propagating waves are Doppler shifted with different frequencies $\omega_{\pm} = \omega \pm k_L v$. The radiation pressure force is the sum of the two forces separately, namely the atomic force $F_{sc,+}$, parallel to the direction of beam propagation, and the force $F_{sc,-}$, which acts in the direction anti-parallel to the beam. These two force components are represented by dotted and dashed lines respectively, and their sum is represented by the unbroken line. Arbitrary units.

3.2. The Dipole Force

The second force is the so-called *dipole force* (known also as *gradient force* or *optical dipole force*). To correctly represent the lineshape of laser light, it can be considered as a mixture of a large number of electromagnetic modes with slightly different frequencies. Under such irradiation, an atom can thereby absorb a photon from one component and release a photon by stimulated emission into another. The net effect of these absorption-stimulated emission cycles on the atomic gross motion is the dipole force, which, for a two-level atom, is given by

$$\mathbf{F}_{dip} = -\frac{\hbar\Delta}{4} \frac{\nabla\Omega^2(\mathbf{R})}{\Delta^2(\mathbf{R}, \mathbf{V}) + \Omega^2(\mathbf{R})/2 + \Gamma^2/4}. \quad (22)$$

For blue detuning ($\omega_L > \omega_0, \Delta > 0$), this force repels atoms from high-intensity regions, while for red detuning ($\omega_L < \omega_0, \Delta < 0$), atoms are attracted towards high intensities. By comparing Equation (20) with Equation (22), we observe that the dipole force increases in intensity, while the scattering force saturates at a maximum value.

The dipole force is a conservative force which may be regarded as resulting from an optical dipole potential given by

$$U_{dip} = \frac{\hbar\Delta}{2} \ln\left(1 + \frac{\Omega^2(\mathbf{R})/2}{\Delta^2(\mathbf{R}, \mathbf{V}) + \Gamma^2/4}\right). \quad (23)$$

In the case of very large detuning, the optical dipole potential assumes the approximate form $U_{dip} = \hbar\Omega^2/\Delta$. The dipole force and its associated potential are responsible for the trapping of atomic motion. Using vortex-structured light, red detuning can be deployed for trapping atoms in an annular high-intensity region, whereas blue detuning enables trapping in the counterpart intensity minima. Such traps can capture single atoms or up to several million in trapping areas with dimensions ranging from a few micrometers to several millimeters.

One of the most significant applications of dipole trapping is found in the generation of *optical lattices*. These are light fields whose intensity structure exhibits periodicity in one, two, or three dimensions, resulting in a landscape of optical dipole traps resembling the structure of crystalline lattices. Here, optical vortices provide for the generation of optical lattices with cylindrical symmetries such as the *optical Ferris wheel* [139] and *helical optical tubes* [70,72].

Although the scattering force is not conservative, due to its dissipative nature, it can be combined with spatially varying magnetic fields created by current-carrying coils to form a robust atom-trapping mechanism. Devices based on this principle, known as the *magneto-optical trap* (MOT), represent the most widely used form of trap in atomic physics laboratories.

Similarly, the conservative dipole force can be exploited in so-called *high-intensity Sisyphus cooling* [140] (in contrast to a polarization mechanism discussed in the following Section 3.3). This effect is experienced by blue-detuned atoms, moving in a high-intensity standing laser beam, with a velocity large enough to travel over several laser wavelengths during its lifetime in the excited state Γ^{-1} but also low enough to neglect non-adiabatic transitions between atom field-dressed states. The decelerating motion can be interpreted on the dressed atom basis as the result of a Sisyphus effect, where the atom moving in an optical potential runs uphill more frequently than downhill.

3.3. Cooling Forces Arising from Polarization Gradients

Soon after the experimental confirmation of Doppler cooling, new experimental data revealed clear violations of the Doppler limit. Further theoretical research confirmed the existence of a polarization-based mechanism for slowing atomic motion, called *polarisation gradient cooling* or simply *Sisyphus cooling*. This mechanism is based on light, whose polarization varies in space. The interaction of this light field with the atom entails electronic transitions involving three atomic levels, not two, as in the previous mechanisms. The result is again a continuous, repetitive cycle of kinetic energy loss from the atom, as the atom climbs potential hills, leading to a very strong deceleration of its motion. When the polarization gradients are formed by two counter-propagating laser beams with mutually orthogonal polarization (the so called *lin \perp lin* configuration), the damping force is given by $F_{lin \perp lin} = -av$ with $a = -3\hbar k^2 \Delta / \Gamma$, ($\Delta = \omega_L - \omega_0 < 0$). When the polarization gradients are formed by two counter-propagating laser beams with opposite circular polarization (the so called $\sigma^+ - \sigma^-$ configuration), the damping force is given by $F_{\sigma^+ - \sigma^-} = -bv$ with $b = -120\hbar k^2 \Delta / \Gamma / [17(5\Gamma^2 + 4\Delta^2)]$, ($\Delta < 0$) [141].

4. Forces in Optical Tweezers

An experiment conducted in 1970 by Ashkin, which eventually led to their shared award of the 2018 Nobel Prize in Physics, represents the birth of the powerful, and now widely used, technique known as *optical tweezers*. In the original experiment, Ashkin succeeded in manipulating the motion of tiny rubber beads using a laser beam [57]. In the form we now know it, the instrumentation of optical tweezers was fully devised by Ashkin and Chu, who succeeded in both trapping and controllably moving miniature dielectric beads [142]. Nowadays, a very diverse and broad range of applications in biology, micromechanics, modelling of thermodynamic systems, colloidal systems, and lab-on-a-chip microchemistry are based on the use of optical tweezers [84].

Optical tweezers have the capacity to control the motion of micro- and nanoparticles. When the dimensions of such a particle are smaller than the wavelength of laser light (the so called *Rayleigh regime*), its motion can be attributed to forces associated with the following relation [143]:

$$\mathbf{F} = \mathbf{F}_{grad} + \mathbf{F}_{diss}. \quad (24)$$

The first force, \mathbf{F}_{grad} , is the the gradient force given by Equation (19). It is a conservative force responsible for confinement in optical tweezers, and it arises from the potential energy of a dipole in the electric field. The second force, \mathbf{F}_{diss} , is given by

$$\mathbf{F}_{diss} = \frac{1}{c}\sigma_{ext}\mathbf{S}(\mathbf{r}) - \frac{1}{2}\sigma_{ext}c\nabla \times \mathbf{s}, \quad (25)$$

where $\mathbf{S} = \frac{1}{2}\Re\{\mathbf{E} \times \mathbf{B}^*\}$ is the real part of the time-averaged Poynting vector, $\mathbf{s} = i\frac{\epsilon_0}{2\omega}\Im\{\mathbf{E} \times \mathbf{E}^*\}$ is the time-averaged spin density of the incoming wave, and c is the speed of light. The quantity $\sigma_{ext} = \frac{k\alpha''}{\epsilon_0}$ is the extinction cross section, and α'' is the imaginary part of the complex polarizability.

It will be evident, on comparing the expression given in Equation (19) with the one in Equation (25), that there is a significant difference. In fact, the former formula is a special case of the latter, being applicable to point-like particles—as we considered an atom when calculating radiation forces. In that regime, as has been extensively discussed, the scattering force is directly proportional to the Poynting vector [144,145].

In the present, more general formulation, the form of the dissipative force shows that it is not simply proportional to the Poynting vector whenever the spin density of the field has a non-vanishing curl. This term has been frequently associated with a third force component [146–148], but it has been shown that it does not play a role in the radiation force. Indeed, the contribution of the Poynting vector can be broken down into an orbital part and a spin part; the spin part is equal and opposite to the spin curl term and cancels it [143,149,150]. In this case, $\mathbf{F}_{diss} = \mathbf{F}_{orbital}$, with $\mathbf{F}_{orbital} = \frac{1}{c}\sigma_{ext}\mathbf{S}(\mathbf{r}) - \frac{1}{2}\sigma_{ext}c\nabla \times \mathbf{s}$. The canonical orbital momentum density alone is responsible for the radiation pressure force.

The two non-conservative components of the radiation pressure force owe their origin to the time-averaged momentum, which in free space can again be separated into two parts, one independent of the polarization and the other dependent upon a degree of circular polarization. The theoretical justification of this distinction rests upon Belifante's symmetrization of the canonical stress energy tensor, which makes the results gauge independent. The part of the dissipative force which depends on the spin-curl density is zero in homogeneous fields and emerges when spatial inhomogeneities of polarization and intensity are present [145]. This force has recently been experimentally measured in an experiment involving circularly polarized evanescent fields [151]. When optical vortex light scatters from a rotating surface, a rotational Doppler effect enables the axis of rotation to be identified [152].

5. Optical Binding

When more than one particle is trapped in a Gaussian laser beam, in an element of the focal region over which the beam has approximately constant intensity, optically induced inter-particle forces arise in addition to the individual trapping forces. Their effect is to establish a potential energy landscape that can hold the particles in a stable, non-contact configuration. First observed by Burns et al. [126], the effect now commonly known as optical binding was first predicted through quantum electrodynamical methods by Thirunamachandran [125]. The effect was later more fully formulated by Bradshaw and Andrews [107], first applied to carbon nanotubes [153] and then extended to multiparticle arrays [98]. Modelling the corresponding Mie scattering effect for larger particles in a trapping cavity leads to results of similar form [127]. Further studies have also identified the additional, optically induced torques that arise when the trapped particles are anisotropic, markedly so in the case of nanowires [128]. The optically induced pair potential is a damped, slowly oscillating function of distance, as illustrated in Figure 2.

Pair trapping usually occurs when the separation between the two particles is at the closest absolute minimum—near to, but not exactly equal to, the optical wavelength. Displacements from this equilibrium position are subject to restoring forces that depend on whether the particles align along the axis (longitudinal binding) or in a plane perpendicular

to it (transverse binding). For particles of polarisability α trapped by a laser of wavenumber k and degree of first order coherence $g_{12}^{(1)}$ across the interparticle divide, [35] the dependence of the forces on their separation R emerges from the energy expressions as follows:

$$F_{binding}^{\parallel}(R) = \frac{\alpha^2 |\mathbf{E}^2| g_{12}^{(1)}}{4\pi\epsilon_0 R^4} \{3 \cos(2kR) - kR \sin(2kR) - 3k^2 R^2 \cos(2kR) - 2k^3 R^3 \sin(2kR)\}, \quad (26)$$

$$F_{binding}^{\perp} = \frac{\alpha^2 |\mathbf{E}^2| g_{12}^{(1)}}{2\pi\epsilon_0 R^4} \{3 \cos(kR) + 3kR \sin(kR) - k^2 R^2 \cos(kR)\}. \quad (27)$$

The pair potential itself can be controllably cancelled by another beam entering a trapping cavity from the side [154]—a technique that also enables the cancellation of dipole forces in Bose–Einstein concentrates [130] and cold atoms [131]. The wider capacity to collectively control optically trapped particle arrays is also a topic of great current interest [100,132].

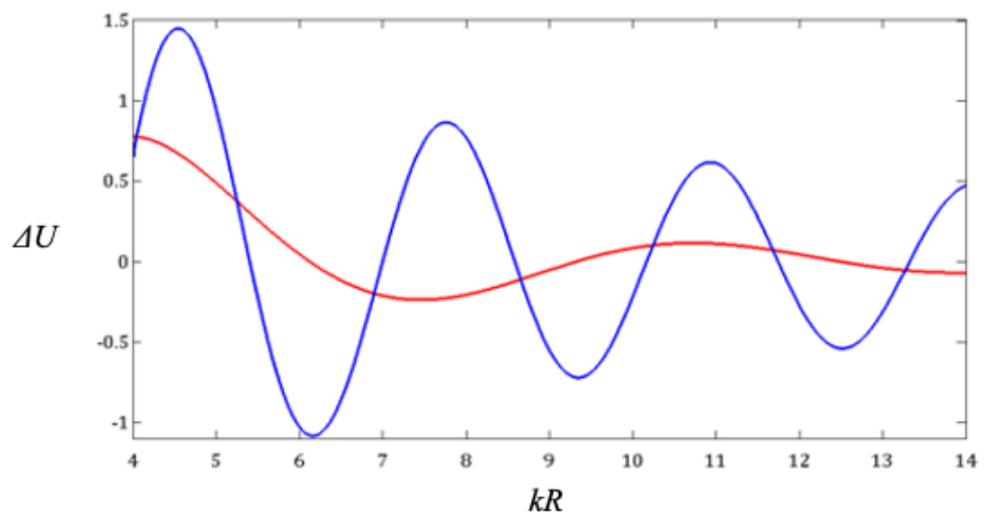


Figure 2. Graph of potential energy, ΔU (arbitrary scale), against kR , where k is the wavenumber, and R the interparticle separation. Axial displacement (blue line); transverse displacement (red line). Reproduced with permission from D.L. Andrews and D.S. Bradshaw, *Optical Nanomanipulation*, second edition (Institute of Physics, Bristol, UK, 2022).

6. Chiral Optical Forces

Across the field of microscale optics, there has recently been a resurgence of interest in the subject of chirality, which concerns entities with a distinct handedness. The simplest methods for studying chiral materials employ light that is itself chiral, usually having a left- or right-handed circular polarization associated with integer spin angular momentum. The local optical chirality density c , defined as

$$\chi = \frac{1}{2} \left\{ \epsilon_0 \mathbf{E} \cdot (\nabla \times \mathbf{E}) + \frac{1}{\mu_0} \mathbf{B} \cdot (\nabla \times \mathbf{B}) \right\}, \quad (28)$$

proves to be directly proportional to a local difference in the numbers of left- and right-handed photons [109,155]. Additionally, beams of light with an azimuthal phase (optical vortices), which convey a different, separable orbital form of angular momentum, represent additional potential for chiral discrimination. In either case, a response according to handedness depends on symmetry breaking through the interference of multipolar interactions of opposite parity [156]. Nearly always, such effects are dominated by an interference of

amplitudes based entirely on electric dipole (E1) interactions, with others in which one electric dipole (odd spatial parity) is substituted by either a magnetic dipole (M1) or electric quadrupole (E2; both are of even spatial parity). Together, these relate to the leading and first-order correction terms in the minimal coupling representation [157,158]. For example, the polarization now emerges in extended form from Equation [131]:

$$\mathbf{P} = \alpha : \mathbf{E} + \mathbf{A} : \nabla \mathbf{E} + \mathbf{G} : \mathbf{B}, \quad (29)$$

where the $E1^2$ polarisability α must now be treated as a second-rank tensor, since a chiral material is necessarily anisotropic. The mixed electric dipole–electric quadrupole $E1E2$ analogue of the polarisability is a third-rank tensor \mathbf{A} which senses the electric field gradient, and its $E1M1$ analogue, commonly represented as \mathbf{G} , engages both the electric and the magnetic field.

There are numerous, well-established spectroscopic methods to achieve chiral *discrimination*, with optical mechanisms underpinned by the framework of theory described above. The achievement of enantiomeric *separation* by optical means is much more challenging. At the molecular level, chirally differential forces are typically of femtonewton magnitude. Most chiral molecules of commercial interest are sizeable (10–100 atoms or more), and in the solution phase, such forces have negligible effect when competing with stochastic Brownian motion. This is a major obstacle; any potential benefit there might appear to be of working in the gas phase is obviated by a correspondingly low vapour pressure and an unrealistically small amount of product. Nonetheless, copious amounts of theory work have been carried out in the search for a usable scheme; a recent review by Genet provides a useful summary [119].

In principle, there is indeed a potential to differentially control the motions of chiral particles using beams of circularly polarized light [102,109,159], but the effects are always small. A variety of more recent and more fanciful schemes have also been proposed to effect chiral separation using beam combinations, but in many cases, the direction of separation reverses within an optical cycle, and they are, accordingly, unrealistic for commercial application [160]. Exploiting the structure of a vortex beam might also enable chiral differentiation to be achieved by exploiting the associated optical gradient forces [161]. Such effects are, in principle, possible even with unpolarized light if the vortex beam is sharply focused, where a small degree of separation into regions of opposite helicity can arise [162]. Again, differential forces occur only in these discrete regions of space, since the volume integral of the optical chirality density necessarily vanishes. The discriminatory force on particles whose dimensions exceed the scale of these regions, or whose thermal motions traverse them, will accordingly disappear.

7. Conclusions

A rich variety of optical force experiments has been devised since the pioneering work of Ashkin in the early days of laser science. The basis for much of the corresponding theory had been laid long before, mostly in connection with large-scale objects, using the classical theory of electromagnetic radiation. The arrival of laser sources afforded an opportunity to tackle some of the more intricate details of the light–matter interaction in terms of the quantum theory of light. This article has accordingly been written in honour of the recently departed Rodney Loudon [163], for his pioneering and inspirational work on this, amongst many other, foundational aspects of quantum optics.

Since the early days, theory has often raced ahead of experiment and prompted investigations that only became possible with the development of new kinds of optical components—the spatial light modulator in the case of structured light, for example. Now, experimental validation awaits many of the most recent predictions. Always, there is the tantalizing prospect of devising methods that can be scaled up for practical applications, aiming to match the outstanding successes of optical tweezer technology. The capability to manoeuvre microscale and nanoscale particles by non-contact means remains the key attraction of laser-based optical forces.

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