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Manipulating particles with light: radiation and gradient forces

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Abstract

The manipulation of matter with electromagnetic radiation is a capacity that has been known for over a century. However, the prominence of such optical effects only grew rapidly following the invention of optical tweezers in the 1980s. While both the original theory and the early trapping techniques are based on the radiation force, optical tweezing uses the gradient force. This paper aims to differentiate between these two clearly distinct types of optical forces, which are sometimes confused in the literature. We also discuss three completely separate forms of optical torque that can be applied to a particle, also due to an electromagnetic field. These involve the transfer of either spin or orbital angular momentum from the beam to the particle, depending on the character of the light, or the often overlooked alignment effect that can act on a cylindrical particle due to a gradient force.

Keywords: optical trap, optical tweezers, optical force, optical manipulation, classical electrodynamics, quantum electrodynamics, photonics

(Some figures may appear in colour only in the online journal)

1. Introduction

The knowledge that matter can be manipulated by light was first put on a firm theoretical basis in the late 19th century. Maxwell and Bartoli [1, 2] determined that light carries linear momentum, which can be transferred to a particle—and as a result, the particle trajectory can be altered by the presence of the light. In 1901, experimental confirmation was given by

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Lebedev [3], and also by Nichols and Hull [4]. Despite this major breakthrough, it was not until the arrival of the laser in the 1960s that production of such a *radiation force* with experimentally meaningful magnitude became possible. An experiment by Ashkin was the first to reveal that laser beams could controllably manipulate particles of matter through the radiation force [5]. However, the associated levels of light intensity—unattainable without the narrow beam-width of a laser—can lead to another previously unknown form of force. This is known as the *gradient force*, and it is the basis for the *optical tweezer* technique [6].

The rapidly burgeoning research literature in this area has unfortunately become replete with an unusual amount of confusion. There are wide variations in terminology, and classical descriptions of theory are frequently vague or sketchy. In some instances research papers will occasionally speak of radiation forces when they mean gradient forces, or *vice-versa*, or may not even distinguish between the two different forces at all. For example, a highly cited paper on optical pulling forces—which are clearly related to radiation forces—provide expressions that seemingly correspond to the gradient force [7]. Such issues of confusion can arise due to a lack of clarity over the nature of the gradient to which reference is being made. Usually, 'gradient force' is understood to relate to a spatially inhomogeneous *intensity*—but if the term were to be applied to *field* gradients then any interaction involving a multipole of higher order than the dipole would be a candidate for the term—which would then misleadingly apply to optical beams of any structure.

It is indeed hard to avoid the conclusion that the authors of some fine experimental studies are unsure of the mechanisms responsible for their observations. Even when such studies are supplemented by computational simulations, these are generally based on classical formulations that are not always appropriate, having no regard to the discrete nature of the radiation responsible for mechanical effects. By providing an explicitly photon-based perspective it is the aim of this article to bring more clarity to the subject. We anticipate that this will make it easier to explain the origins of the wide variety of forces and torques that can be produced by light. The central aim is to differentiate between the separate forms of optical force: (in brief) one involves the 'push' of the irradiated particle in the propagation direction of the light, and the other consists of an attraction of the particle to the beam centre. The theory behind the optical forces that act on nano- rather than micro-particles will be discussed in the main; however, microscale systems are mentioned throughout. We hope that these discussions will help assist the task of overriding any confusion that sometimes arises in the literature.

First, in section 2, the attributes of the electromagnetic radiation in the free field are presented. Here we outline, without resorting to detailed derivations, the three mechanical quantities that could be delivered by a photon; first offering the classical representations familiar to some readers and then the quantum equivalents. This is followed by the introduction of a framework to categorise light–matter interactions, which is then used to assist the description of the radiation and gradient forces (section 3). Section 4 then addresses optical torque, including an analysis of the often overlooked torque that the gradient force can exert on a cylindrical particle. This is completely separate in its influence from two other forms of torque—corresponding to the radiation force—that relate to the spin angular momentum of circularly polarised light or the orbital angular momentum of a twisted beam; the latter is connected to the idea of an *optical spanner* [8]. Each of these torques is fully analysed, and the paper then concludes in section 5 with a discussion.

2. Theoretical foundation

2.1. Light in the free-field

A photon is known to convey three mechanical quantities: energy, linear momentum and angular momentum. Energy, as a scalar, cannot contribute any sort of directionality onto a particle as a result of light absorption—or indeed any other optical process. In contrast, the directionality of the optical linear and angular momenta can indeed be conferred to a particle. This is the basis for the radiation pressure that, for a beam of sufficient intensity, may produce linear forces and angular forces (torques). The latter may also be produced via the gradient force, as a consequence of a non-uniform beam intensity distribution that produces a local potential energy landscape (*vide infra*).

Here, we introduce the key electromagnetic variables and the main equations that quantify these features. A classical Fourier series representation can be written for the generalised position- and time-dependent electric and magnetic free-fields, $\mathbf{E}(\mathbf{r}, t)$ and $\mathbf{B}(\mathbf{r}, t)$ respectively, associated with electromagnetic radiation [9],

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}^{(+)}(\mathbf{r}, t) + \mathbf{E}^{(-)}(\mathbf{r}, t); \quad \mathbf{B}(\mathbf{r}, t) = \mathbf{B}^{(+)}(\mathbf{r}, t) + \mathbf{B}^{(-)}(\mathbf{r}, t), \tag{1}$$

where each field is written as a sum of two mode-summed terms. These are known as the positive and negative frequency parts of the fields, respectively. The signs in the superscripts relate to spatial phase factors that feature in the explicit Fourier expansions: for example, the term $\mathbf{E}^{(+)}(\mathbf{r}, t)$ (known as the *analytic signal*) is given by;

$$\mathbf{E}^{(+)}(\mathbf{r}, t) = \mathrm{i}\sum_{\ell} \mathbf{e}_{\ell}\mathscr{E}_{\ell}\alpha_{\ell}(t)\mathrm{e}^{\mathrm{i}(\mathbf{k}_{\ell}\cdot\mathbf{r})}, \tag{2}$$

where the Fourier component with index ℓ relates to a set of four numbers relating to the wave-vector $\mathbf{k}_{\ell}$ (three Cartesian components) and polarisation unit vector $\mathbf{e}_{\ell}$; $\alpha_{\ell}(t)$ is a normal variable dependent on the positive form of the phase factor and $\mathscr{E}_{\ell}$ is a mode constant; $\mathbf{E}^{(-)}(\mathbf{r}, t)$ is the complex conjugate of the expression given by equation (2). The Fourier representations of the two components of $\mathbf{B}(\mathbf{r}, t)$ have a similar form. To satisfy Maxwell's equations, the time-dependent factor $\alpha_{\ell}(t)$ has to carry a temporal phase factor $\exp(-\mathrm{i}\omega t)$. Thus, individual modal components of the Fourier series can be regarded as representing radiation of angular optical frequency $\omega = ck$, propagating with a wave-vector $\mathbf{k}$.

For analogous expressions in the quantum formulation, i.e. where the electric and magnetic fields are quantised, the factor α_{ℓ} and its conjugate are promoted to operator status —signified here, and henceforth, by the carat symbol—to obtain [10],

$$\hat{\mathbf{E}}(\mathbf{r}) = \hat{\mathbf{E}}^{(+)}(\mathbf{r}) + \hat{\mathbf{E}}^{(-)}(\mathbf{r}) = \mathrm{i}\sum_{\mathbf{k},\eta}\left(\frac{\hbar ck}{2\varepsilon_0 V}\right)^{\frac{1}{2}}\{\mathbf{e}^{(\eta)}(\mathbf{k})\hat{a}^{(\eta)}(\mathbf{k})\mathrm{e}^{\mathrm{i}(\mathbf{k}\cdot\mathbf{r})} - \bar{\mathbf{e}}^{(\eta)}(\mathbf{k})\hat{a}^{\dagger(\eta)}(\mathbf{k})\mathrm{e}^{-\mathrm{i}(\mathbf{k}\cdot\mathbf{r})}\} \tag{3}$$

$$\hat{\mathbf{B}}(\mathbf{r}) = \hat{\mathbf{B}}^{(+)}(\mathbf{r}) + \hat{\mathbf{B}}^{(-)}(\mathbf{r}) = \mathrm{i}\sum_{\mathbf{k},\eta}\left(\frac{\hbar k}{2\varepsilon_0 cV}\right)^{\frac{1}{2}}\{\mathbf{b}^{(\eta)}(\mathbf{k})\hat{a}^{(\eta)}(\mathbf{k})\mathrm{e}^{\mathrm{i}(\mathbf{k}\cdot\mathbf{r})} - \bar{\mathbf{b}}^{(\eta)}(\mathbf{k})\hat{a}^{\dagger(\eta)}(\mathbf{k})\mathrm{e}^{-\mathrm{i}(\mathbf{k}\cdot\mathbf{r})}\}, \tag{4}$$

where the summation is again taken over each mode $(\mathbf{k}, \eta)$, in which η labels the polarisation state, and V is an arbitrary quantisation volume—a notional region within which the system of interest resides. The Hermitian conjugate pair of operators $\hat{a}^{(\eta)}(\mathbf{k})$ and $\hat{a}^{\dagger(\eta)}(\mathbf{k})$ signify photon annihilation and creation, respectively, i.e. for each mode they act to reduce or increase by

one the number of photons present in the system; the electric and magnetic polarisation vectors of each photon, denoted by $\mathbf{e}^{(\eta)}(\mathbf{k})$ and $\mathbf{b}^{(\eta)}(\mathbf{k}) = \mathbf{e}^{(\eta)}(\mathbf{k}) \times \mathbf{k}$ (overbars signifying their complex conjugates), also appear in the above expressions. In the quantum formalism, $\hat{\mathbf{E}}(\mathbf{r})$ and $\hat{\mathbf{B}}(\mathbf{r})$ are not dependent on time since, in the assumed Schrödinger representation, any time dependence is held within the state vector.

The classical Hamiltonian of the radiation field, H_{rad}, entails an integral of the energy density over volume, namely [11];

$$H_{\text{rad}} = \frac{\varepsilon_0}{2} \int_V (|\mathbf{E}(\mathbf{r}, t)|^2 + c^2 \, |\mathbf{B}(\mathbf{r}, t)|^2) \mathrm{d}^3 r. \tag{5}$$

On substitution of the quantum field operators, i.e. equations (3) and (4), into this expression—where V again acquires the significance of a quantisation volume—the following quantum Hamiltonian operator is determined;

$$\hat{H}_{\text{rad}} = \sum_{\mathbf{k},\eta} \left(\hat{N}^{(\eta)}(\mathbf{k}) + \frac{1}{2} \right) \hbar\omega, \tag{6}$$

where the dependence of ω on $\mathbf{k}$ is left implicit; $\hat{N}^{(\eta)}(\mathbf{k}) = \hat{a}^{\dagger(\eta)}(\mathbf{k})\hat{a}^{(\eta)}(\mathbf{k})$ is the *number operator*, which returns the number of photons when applied to a number (Fock) state. Moreover, for the ground state of the radiation, $\frac{1}{2}\hbar\omega$ represents the zero point energy, associated with vacuum fluctuations that are present even in the absence of radiation. On inspection of equation (6), the energy of a single photon is determined as $\hbar\omega$.

The classical *linear* momentum of the electromagnetic radiation, $\mathbf{P}_{\text{rad}}$, involves an integral of the Poynting vector over V;

$$\mathbf{P}_{\text{rad}} = \varepsilon_0 \int_V (\mathbf{E}(\mathbf{r}, t) \times \mathbf{B}(\mathbf{r}, t)) \mathrm{d}^3\mathbf{r}, \tag{7}$$

while the total *angular* momentum of the radiation, $\mathbf{J}_{\text{rad}}$, is proportional to an integral of the cross product between the Poynting vector and displacement, namely;

$$\mathbf{J}_{\text{rad}} = \varepsilon_0 \int_V \mathbf{r} \times (\mathbf{E}(\mathbf{r}, t) \times \mathbf{B}(\mathbf{r}, t)) \mathrm{d}^3\mathbf{r}. \tag{8}$$

With the use of simple vector identities, the latter expression can be re-written as a sum of terms relating to the spin, $\mathbf{S}_{\text{rad}}$, and orbital angular momentum, $\mathbf{L}_{\text{rad}}$, as signified by the respective terms in the following;

$$\mathbf{J}_{\text{rad}} = \mathbf{S}_{\text{rad}} + \mathbf{L}_{\text{rad}} = \varepsilon_0 \left(\int_V \mathbf{E}(\mathbf{r}, t) \times \mathbf{A}(\mathbf{r}, t) + E_i(\mathbf{r}, t)(\mathbf{r} \times \nabla) A_i(\mathbf{r}, t) \mathrm{d}^3\mathbf{r} \right). \tag{9}$$

Here, $\mathbf{A}(\mathbf{r}, t)$ is the electromagnetic vector potential [12] that is related to the electric and magnetic fields via $\mathbf{E} = -\partial \mathrm{A}/\partial \mathrm{t} - \nabla\Phi$ and $\mathbf{B} = \nabla \times \mathbf{A}$, respectively, where ∇ denotes a differential with respect to $\mathbf{r}$ and Φ is the electromagnetic scalar potential. In the latter term of equation (9), the repeated index i represents summation (also known as an Einstein summation [13]) over any frame of Cartesian coordinates. It is notable that the form of separation in equation (9) is not unique, since the electromagnetic vector potential is not gauge invariant. In the Coulomb gauge [14, 15] used in this work, the decomposition of $\mathbf{J}_{\text{rad}}$ into $\mathbf{S}_{\text{rad}}$ and $\mathbf{L}_{\text{rad}}$ is achievable on the assumption that the optical beam has paraxial form—i.e. it is subject to the same conditions as apply for the small-angle approximation in Gaussian optics. The decomposition of total angular momentum into two terms is much more difficult for non-paraxial beams

The quantum framework involves the following explicit mode expansion of the vector potential:

$$\hat{\mathbf{A}}(\mathbf{r}) = \sum_{\mathbf{k},\eta}\left(\frac{\hbar}{2\varepsilon_0 ckV}\right)^{\frac{1}{2}}\{\mathbf{e}^{(\eta)}(\mathbf{k})\hat{a}^{(\eta)}(\mathbf{k})\mathrm{e}^{\mathrm{i}(\mathbf{k}\cdot\mathbf{r})} - \bar{\mathbf{e}}^{(\eta)}(\mathbf{k})\hat{a}^{\dagger(\eta)}(\mathbf{k})\mathrm{e}^{-\mathrm{i}(\mathbf{k}\cdot\mathbf{r})}\}. \tag{10}$$

By inserting the field operators (3), (4) and (10) into the corresponding classical expressions $\mathbf{P}_{\mathrm{rad}}$, $\mathbf{S}_{\mathrm{rad}}$ and $\mathbf{L}_{\mathrm{rad}}$ (as required) their quantum equivalents are produced. Remarkably simple expressions for the linear and angular momenta then emerge, readily related to photon quantities. First, it is found that;

$$\hat{\mathbf{P}}_{\mathrm{rad}} = \sum_{\mathbf{k},\eta}\hat{N}^{(\eta)}(\mathbf{k})\hbar\mathbf{k}, \tag{11}$$

in which the zero-point momentum for each mode, ½$\hbar\mathbf{k}$, vanishes due to each $\mathbf{k}$ in the summation having a matching –$\mathbf{k}$ (compare with the zero-point energy, which persists because energy is solely a positive quantity). From this expression, we determine that the linear momentum of each photon is $\hbar\mathbf{k}$.

Next, we can observe that radiation states of pure circular polarisation are eigenstates of the operator for quantum spin, $\mathbf{S}_{\mathrm{rad}}$. This is consistent with an angular momentum of $\pm\hbar$ per photon with the sign determined by handedness; the vector quantity directed along $\mathbf{k}$ is thus expressible as $\pm\hbar\mathbf{u}_{\mathbf{k}}$, where $\mathbf{u}_{\mathbf{k}}$ represents a unit vector of $\mathbf{k}$. This spin feature is, therefore, conveyed by the following;

$$\hat{\mathbf{S}}_{\mathrm{rad}} = \sum_{\mathbf{k}}\{\hat{N}^{(L)}(\mathbf{k}) - \hat{N}^{(R)}(\mathbf{k})\}\hbar\mathbf{u}_{\mathbf{k}}, \tag{12}$$

with the mode summation specifically cast in terms of circular polarisations, either left- or right-handed light as denoted by the superscript L and R. This expression indicates, as might be expected, that the spin-angular momentum operator, $\hat{\mathbf{S}}_{\mathrm{rad}}$, depends solely on the disparity of left- and right-handed photon populations [16, 17]. Alongside this result it also emerges that the orbital angular momentum operator, $\hat{\mathbf{L}}_{\mathrm{rad}}$, is most expediently expressed in terms of a modal basis associated with a helical wavefront, such as Laguerre-Gaussian modes;

$$\hat{\mathbf{L}}_{\mathrm{rad}} = \sum_{k,\eta,l,p}\hat{N}_{lp}^{(\eta)}(\mathbf{k})l\hbar\mathbf{u}_{\mathbf{k}}, \tag{13}$$

in which the four degrees of freedom are the magnitude of a wave-vector k, polarisation η, and the indices l and p as integers that designate the order and degree of the corresponding Laguerre polynomial. The former index is often known as the *topological charge*; the latter is the radial index that represents the number of radial nodes, $p + 1$, within the transverse field distribution of the beam. In passing, it is useful to note that by casting the sum over η explicitly in terms of a basis comprising left- and right-handed circular polarisations, we arrive at [18];

$$\hat{\mathbf{L}}_{\mathrm{rad}} = \sum_{k,l,p}\{\hat{N}_{lp}^{(L)}(\mathbf{k}) + \hat{N}_{lp}^{(R)}(\mathbf{k})\}l\hbar\mathbf{u}_{\mathbf{k}}, \tag{14}$$

which forms a neat counterpart to the spin angular momentum as given by equation (12). The above expressions indicate that the orbital angular momentum has no connection with any spin helicity associated with polarisation [19]. Furthermore, we can determine that a free field photon endowed with topological charge has an orbital angular momentum of magnitude $l\hbar$.

2.2. Light–matter interactions

At this point, only the electromagnetic radiation on its own (i.e. the free field) has been examined. We now consider a nanoscale system in which light engages with matter. A

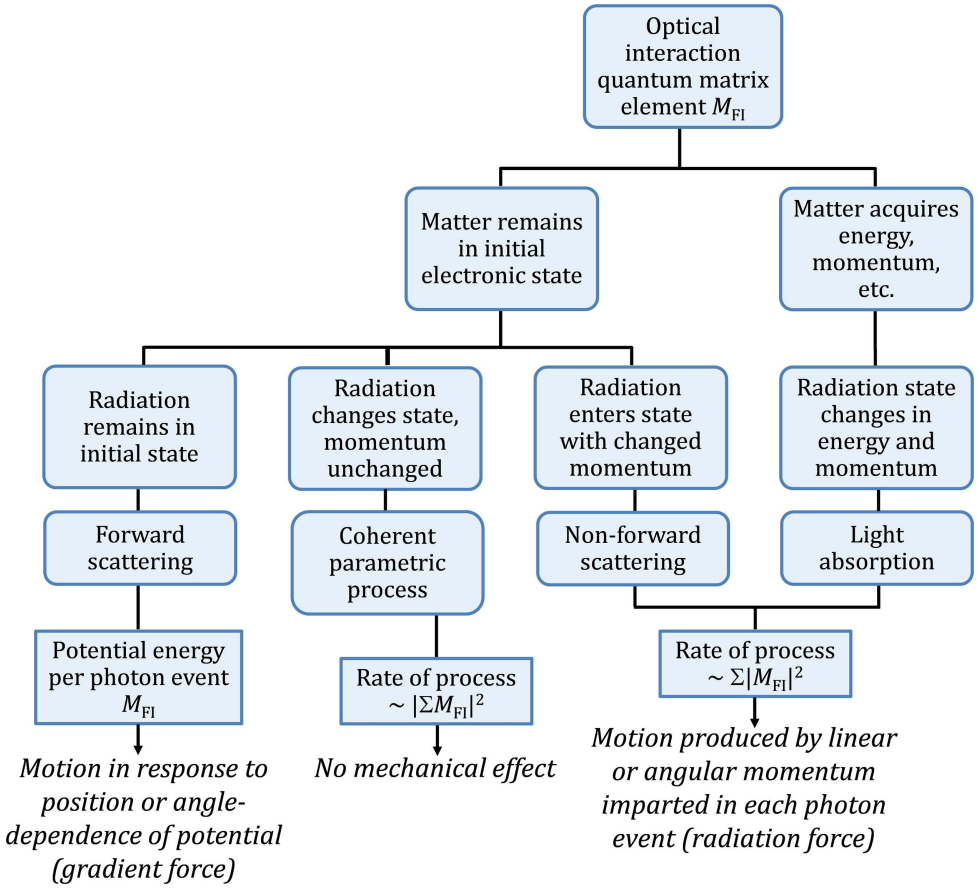

Figure 1. Flowchart for a nanoscale system that exhibits essential differences in implementation of the matrix element. The different pathways of interpretation are determined by considering any change in the quantum state of the matter and radiation; the relevant mechanism and its measurable are then decided.

conventional starting point is the definition of a total Hamiltonian $\hat{H}$, expressed as the sum of three parts;

$$\hat{H} = \hat{H}_{\text{part}} + \hat{H}_{\text{rad}} + \hat{H}_{\text{int}}, \tag{15}$$

where $\hat{H}_{\text{part}}$ and $\hat{H}_{\text{int}}$ are the particle and interaction Hamiltonian, respectively, and the interaction acts as a perturbation on both particle and radiation states. Theory, based on this perturbation, is then developed from the following general expression for the quantum matrix element, M_{FI}, for progression from an initial system state $|I\rangle$ to a final state $|F\rangle$;

$$M_{FI} = \sum_{q=0}^{\infty} \langle F | \{ \hat{H}_{\text{int}} (\hat{T}_0 \hat{H}_{\text{int}})^q \} | I \rangle. \tag{16}$$

Here $\hat{T}_0$, known as a *resolvent operator*, is given by $\hat{T}_0 \approx (E_I - \hat{H}_0)^{-1}$, where the unperturbed system is represented by $\hat{H}_0 = \hat{H}_{\text{mat}} + \hat{H}_{\text{rad}}$ and E_I is the energy of the initial system state.

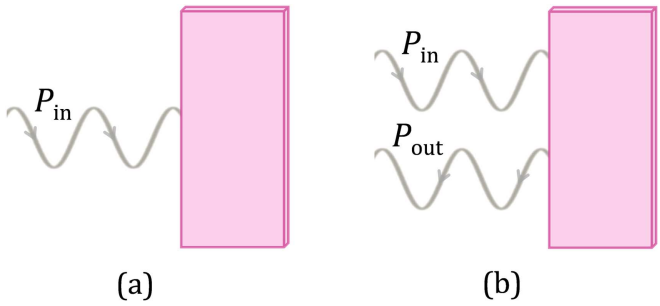

Figure 2. Radiation pressure, determined from the Maxwell–Bartoli expression, applied to a surface due to (a) absorption ($R = 0$) and (b) reflection ($R = 1$): P_{in} and P_{out} represent the radiation pressure due to the incident and reflected light, respectively.

The status and application of a matrix element as given by equation (16) differs according to the nature of the optical process for which it is evaluated. To assist in identifying the correct route for applications that can produce mechanical motion, a flowchart that leads to the appropriate physical observable is presented in figure 1. By suitable use of equation (16), all of the key electrodynamical properties can be derived, and also analytical expressions for the observables associated with optical processes—such as rates, intensities and forces. For example, the $q = 0$ term delivers *transition* dipole moments and oscillator strengths for absorption processes, while the $q = 1$ term gives an explicit expression for polarisability, as we shall see below. The importance of all these factors in the determination of optical forces will emerge in the following.

3. Optical forces

The generation of optical forces may occur via the interaction of the electromagnetic field of light with the charge distribution of a particle. The effect is most obvious under resonance conditions, where a photon energy matches the difference between any two electronic energy levels in the absorber—this is the radiation force. However, such fields may also exert influence under non-resonance conditions that involve short-lived virtual transitions. In this case, where the location of the particle relative to the local distribution of the light is the key, a gradient force may result from the position-dependent energy. Here, the light–matter interactions produce only transient change to the internal electronic energy of the material. In terms of photonics, optical manipulation of neutral particles by a laser beam may, hence, involve two completely separate mechanisms.

3.1. Radiation force

The radiation force, traditionally associated with radiation pressure, relates to the transfer of linear momentum from the electromagnetic radiation to the particle. This can occur either through light absorption, or a change in the direction of the light due the interaction with the particle (non-forward scattering). The classical and quantum descriptions are now outlined, recognising the connection between the essentially classical, macroscopic and more obviously quantum level, nanoscopic forces associated with optical radiation.

For applications on the macroscopic scale the original Maxwell–Bartoli force is readily used, to account for the force that light exerts onto a surface. Since an optical force can be

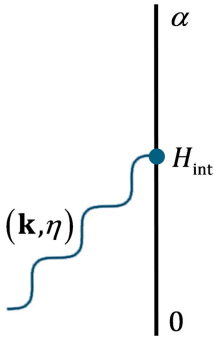

Figure 3. Feynman diagram illustrating the radiation force due to single photon absorption. The blue wavy line represents a photon of mode ($\mathbf{k}$, η), the vertical line symbolises an optical centre that begins in a ground state 0 and finishes in its excited state α, the blue dot denotes the light–matter interaction, labelled by H_{int}, and time travels upwards. Since the initial state $|I\rangle = |n; 0\rangle$ and final state $|F\rangle = |(n-1); u\rangle$ differ, this diagram represents a process in which the physical observable is a rate, delivered from the matrix element using the Fermi rule.

described by both energy per unit distance and momentum per unit time (from Newton's second law) then it follows that optical momentum can be defined as energy divided by the speed of light. The radiation pressure, P, is the rate of change of momentum divided by the irradiated area so that: $P = I\cos^2\theta/c$, where I is the irradiance—the energy arriving at the surface per unit time (input power) per unit area, and θ is the angle between the surface normal and the incident radiation. This expression describes the radiation pressure acting on a perfectly absorbing surface, which is illustrated by figure 2(a). If the surface is perfectly reflecting, then a factor of two is included on the right-hand side of the expression; the latter is explained by the fact that a momentum is exerted by both the incident and reflected light, as shown by figure 2(b). In practice, the pressure delivered to a surface lies somewhere between the two extremes, according to its reflectivity R. Hence, the general Maxwell–Bartoli expression emerges as follows:

$$P = (1 + R)(I/c)\cos^2\theta. \tag{17}$$

At the quantum level, the radiation force when n_ϕ photons are absorbed by one or more particles is determined by multiplying the absorption rate, Γ, by the net linear momentum of the absorbed photons, $n_\phi\hbar\mathbf{k}$. The rate of absorption, for a given wave-vector $\mathbf{k}$ and polarisation η, can accordingly be found by application of the Fermi rule, $\Gamma = (2\pi\rho_F/\hbar)|M_{FI}|^2$, where ρ_F is the density of states and M_{FI} is the matrix element determined from first-order time-dependent perturbation theory [20]. The result emerges from the $q = 0$ term in equation (16) as:

$$M_{FI} = \langle F\,|\hat{H}_{\text{int}}|\,I\rangle = \langle (n-1); u\,|-\hat{\boldsymbol{\mu}}\cdot\hat{\mathbf{E}}(\mathbf{r})|\,n; 0\rangle. \tag{18}$$

At this juncture, it is appropriate to introduce the Feynman diagrammatic representation; the diagram corresponding to equation (18) is shown in figure 3. Using Dirac bracket notation, $|I\rangle = |n; 0\rangle$ and $|F\rangle = |(n-1); u\rangle$ represent the specific initial and final system states, respectively, decomposed into radiation and particle components: the initial state contains n photons and the particle is in the ground state 0, and the final state (which follows photon absorption) comprises n–1 photons and the particle in its excited state u.

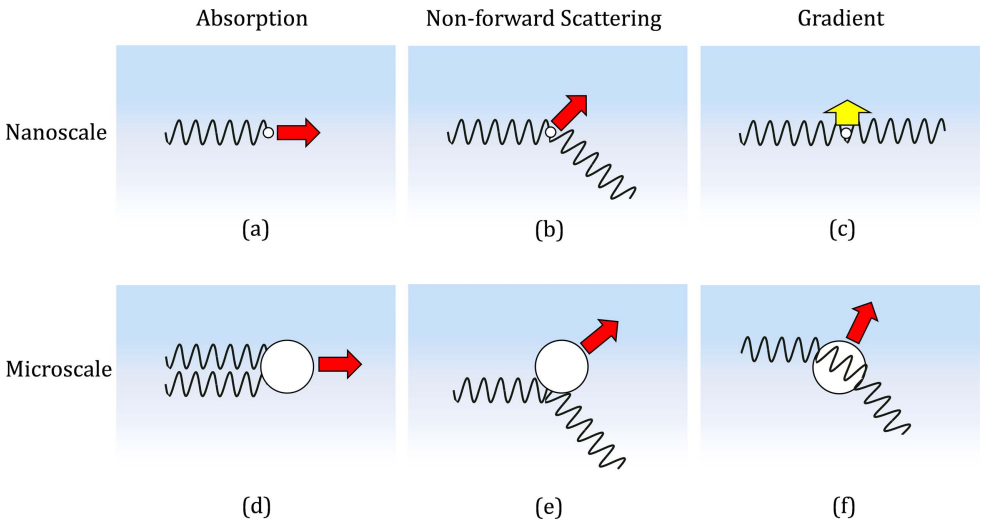

Figure 4. Diagrams indicating the mechanism behind each of three different kinds of laser-induced force (shown by the arrows) acting on a nano- or micro-particle (circle) due to a beam of laser radiation (wavy line) propagating from left to right. Red arrows correspond to a direct transfer of momentum and yellow to an indirect force associated with a position-dependent dynamic Stark effect. The radial distribution of laser beam intensity is represented in blue; the beam axis (the highest intensity part of the beam) is positioned at the top of each diagram and the intensity diminishes below: (a), (d) involve light absorption and (b), (e) non-forward scattering, each producing a radiation force; (c), (f) relate to the gradient force, which is always directed towards the highest intensity region. In (f), due to the intensity profile, more light impinges the upper portion of the microparticle than the lower part, and as a result of refraction the particle experiences a net force upwards.

The structure of equation (18) reveals that it is a change in state of the radiation field that allows for the transfer of momentum from the irradiating light to the particle. Also appearing in equation (18) is the explicit expression for the interaction Hamiltonian, i.e. $\hat{H}_{\text{int}} = -\hat{\boldsymbol{\mu}} \cdot \hat{\mathbf{E}}(\mathbf{r})$ where $\hat{\boldsymbol{\mu}}$ is the electric dipole moment operator that acts on the particle states to produce a *transition* dipole moment $\boldsymbol{\mu}^{u0}$, and $\hat{\mathbf{E}}(\mathbf{r})$ acts on the radiation states. Here, in fact, only the $\hat{\mathbf{E}}^{(+)}(\mathbf{r})$ portion of equation (3) is required since it is this term that includes the photon annihilation operator. Assuming $\mathbf{e}||\boldsymbol{\mu}^{u0}$ and the density of states is $\rho_F = I_\omega V/2\pi c n \hbar 2\omega$ [21], the radiation force is determined as;

$$\mathbf{F} = \frac{n_\phi I_\omega \mathbf{k}}{2c\varepsilon_0 \hbar} \, |\boldsymbol{\mu}^{u0}|^2, \tag{19}$$

where I_ω is the irradiance per unit frequency. In cases where the incident photons are deflected, rather than absorbed, the treatment of theory is a little more intricate and involves second-order perturbation theory; such a treatment is presented in section 4 for a torque based on radiation pressure.

3.2. Gradient force

At the atomic or molecular level, the gradient force involves the production of intensity-dependent internal energy level shifts, associated with the dynamic (ac) Stark effect. In such a case, since the level shifts depend on the intensity of the light, an effective potential energy

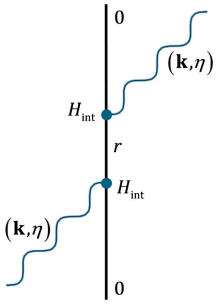

Figure 5. Feynman diagram illustrating the gradient force based on forward-Rayleigh scattering, in which a photon is annihilated and another recreated into the same radiation mode. The optical centre begins and ends in the ground state, traversing through a short-lived virtual state *r*. Since the initial and final state are identical, i.e. $|I\rangle = |F\rangle = |n; 0\rangle$, this diagram represents a potential energy obtained from the real part of the matrix element.

surface is introduced whose magnitude varies with particle position—usually within the cross-section of the trapping beam and which is, thus, related to the transverse field distribution. The gradient force is determined from the spatial derivative of the potential energy; in consequence the particle is attracted to the high intensity part of the trapping beam. The gradient force that acts on any transparent *micro*particle, by contrast, is primarily associated with a different effect, whose mechanism is usually treated using a ray optics approach. Here, a difference in the relative photon flux impinging on the surface of the particle closest to, and furthest from, the beam axis produces correspondingly different rates of deflection. Effectively this produces attraction to the high intensity part of the beam as momentum is transferred from the light to the particle. A complete overview, in diagrammatic form, of each type of optical force when applied to either a nano- or micro-scale particles is given by figure 4.

In terms of classical electrodynamics, the gradient force is conventionally defined in terms of the Lorentz force on a point dipole which, introducing a constant of proportionality (a scalar polarisability) α, is expressed as;

$$\mathbf{F}(\mathbf{r}, t) = \alpha\left(\frac{1}{2}\nabla E(\mathbf{r}, t)^2 + \frac{\mathrm{d}}{\mathrm{d}t}(\mathbf{E}(\mathbf{r}, t) \times \mathbf{B}(\mathbf{r}, t))\right). \tag{20}$$

The second term relates to the Poynting vector—which can again be cast, using equation (1), as a sum of four terms involving $\mathbf{E}^{(+)}(\mathbf{r}, t)$ and $\mathbf{E}^{(-)}(\mathbf{r}, t)$ in cross-products with their magnetic counterparts. It then transpires that the product contributions $\mathbf{E}^{(+)}(\mathbf{r}, t)\mathbf{B}^{(-)}(\mathbf{r}, t)$ and its complex conjugate are explicitly time-independent; the two other terms carry factors $\exp(\pm 2\mathrm{i}\omega t)$, and hence oscillate too rapidly to engage with whole-particle motion. The 'Poynting' term in equation (20) thus disappears, and the time-averaged optical force (denoted below by $\langle\rangle_{\mathrm{T}}$) becomes [22];

$$\langle \mathbf{F}(\mathbf{r})\rangle = \frac{\alpha}{2}\nabla\langle E(\mathbf{r})^2\rangle_{\mathrm{T}}. \tag{21}$$

Here, the electric field is involved in a quadratic response and, therefore, we can conclude that the gradient force is sustained in an oscillating field.

Returning to the quantum theory, contrasting to any force that relates to the radiation pressure, the gradient force does not involve a transfer of momentum (unlike the microscale case) since the initial and final system states are identical and, hence, the input electromagnetic field suffers no change. Here, the position-dependent potential energy, ΔE, is evaluated as an

expectation value (i.e. a matrix element in which the initial and final states are identical) so that the interaction involves photon annihilation and recreation into the same radiation mode. This mechanism is known as forward-Rayleigh (elastic) scattering, as shown in figure 5, wherein the frequency of the trapping beam is non-resonant with any energy level transitions in the particle.

Using second-order perturbation theory, i.e. the $q = 1$ term in equation (16), the following matrix element signifies the evaluation of the expectation value;

$$\Delta E = \mathrm{Re}\, M_{II} = \mathrm{Re}\left\{\sum_R \frac{\langle I|\ \hat{H}_{\mathrm{int}}\ |R\rangle\langle R|\ \hat{H}_{\mathrm{int}}\ |I\rangle}{E_I - E_R}\right\}, \tag{22}$$

where $|I\rangle = |n;\, 0\rangle$ denotes both the initial and final system state and $|R\rangle = |n-1;\, r\rangle$ is an intermediate system state: E is the energy of the state denoted by the subscript. It is important to note that $|R\rangle$ involves a short-lived virtual particle state r, rather than any specific excited state u. Moreover, $\hat{H}_{\mathrm{int}} = -\hat{\boldsymbol{\mu}} \cdot \hat{\mathbf{E}}(\mathbf{r})$ will invoke either term of equation (3) so that either $\hat{\mathbf{E}}^{(+)}(\mathbf{r})$ or $\hat{\mathbf{E}}^{(-)}(\mathbf{r})$ appears in $\hat{H}_{\mathrm{int}}$, depending on whether the operator acts on a photon annihilation or creation event. It is noteworthy that $\hat{H}_{\mathrm{int}}$ could involve additional terms, for example that relate to a *transition* magnetic dipole rather than an electric dipole, although such contributions are very small and they are rarely considered in conventional optics; however, a magnetic dipole interaction is vital in connection with the gradient force of different magnitudes that acts on chiral molecules with opposite handedness [23–25]. Using a beam irradiance (in units of W m^{-2}) given by $I = n\hbar c^2 k/V$, the gradient force is found as;

$$\mathbf{F}(\mathbf{r}) = \left(\frac{\nabla I(\mathbf{r})}{2\varepsilon_0 c}\right)\alpha_{ij}(\omega)\bar{e}_i e_j. \tag{23}$$

Here, the η and $\mathbf{k}$ dependences are suppressed on the unit polarisation vector $\mathbf{e}$, and the polarisability $\alpha_{ij}(\omega)$ emerges, in a complete form, as a second rank property tensor explicitly given by;

$$\alpha_{ij}(\omega) = \sum_r \left\{\frac{\mu_i^{0r}\mu_j^{r0}}{E_{r0} - \hbar\omega} + \frac{\mu_j^{0r}\mu_i^{r0}}{E_{r0} + \hbar\omega}\right\}, \tag{24}$$

where $E_{0r} = E_0 - E_r$. Equation (23) is written in a concise form that again uses the Einstein summation convention (for repeated tensor indices). In any application to three-dimensional space, it simply means that where subscript indices i, j are used to signify x, y or z components in a Cartesian frame of reference, the repetition of an index within a given term signifies an implicit summation over those same three directions. For example, a scalar product $\boldsymbol{\mu} \cdot \mathbf{E}$ may be written as $\mu_i E_i$ and the scalar $\alpha_{ii} = \mathrm{tr}(\alpha)$. It transpires that equation (23) tallies with equation (21) through a rotational average leading to $\langle \alpha_{ij}(\omega)\bar{e}_i e_j\rangle_R = \frac{1}{3}\mathrm{tr}(\boldsymbol{\alpha}) = \alpha$ together with the relation $I = c\varepsilon_0|\mathbf{E}|^2$. This confirms that the derivation of optical force based on photon optics represents the same physics as the more widely known Lorentz force.

4. Optical torque

The torque that may act upon a particle due to the application of a light beam can have three, completely separate forms. The better known types of torque, based on the radiation force, involve the angular momentum of the electromagnetic radiation. Here, the torque results in either the spin of the particle on its axis due to light with *spin* angular momentum, or the orbit around a central point owing to radiation with *orbital* angular momentum. However, there is another form of torque based on the gradient force, which has the capacity to align a

cylindrical particle in a certain direction within the beam. These three types of torque are separately discussed in the following.

4.1. Torque dependent on gradient force

The torque due to the gradient force will *not* successfully act on a spherical particle, since the latter relates to a polarisability tensor that is represented by a diagonal matrix with all three components identical. This, perhaps, explains why this orientational effect is not well-known, since the majority of optical trapping studies use spherical particles. The simplest case of a non-spherical nanoparticle is one with cylindrical symmetry, in which the diagonal of the polarisability matrix has one value, $\alpha_{||}$, for the axial direction, and another, $\alpha_{\perp}$, for each of the other two axes. Here, $\alpha_{||}$ will always have the larger magnitude in a prolate ellipsoid since, in the longer axial direction, the polarisability—which is, basically, a measure of the shift in electron distribution to engender a dipole moment—will correspond to a larger electron shift.

Any such nanotube, sitting initially in an arbitrary angle with respect to the linearly polarised input radiation, will experience a torque that tends to align its long axis with the polarisation plane, at right-angles to the direction of light propagation. However, within a laser beam with a narrow waist, a larger microscale particle with cylindrical symmetry *will* align with the propagation direction of the light, to maximise engagement with the high intensity region [26]. An expression for the nanoscale torque is given by;

$$\boldsymbol{\tau} = (\alpha_{\perp} - \alpha_{||})(E_y\mathbf{i} - E_x\mathbf{j})E_z, \tag{25}$$

where E_x, E_y and E_z are the electric field directed in the x-, y- and z-directions: $\mathbf{i}$ and $\mathbf{j}$ respectively signify x- and y-unit vectors.

4.2. Torque due to spin angular momentum

Another form of optical torque arises when circularly polarised light, which contains spin angular momentum, is applied to a particle (that is again, in practice, usually spherical). This type of torque has been shown to produce an extremely quick particle rotation rate, in fact the world's fastest spinning man-made object is generated by such a torque—an entity so fast that the angular acceleration at the sphere surface is a billion times that of gravity on the Earth's surface [27]. A torque of this form is based on the radiation force, since spin angular momentum is transferred from the light beam to the particle. As such, instead of the transferal of optical linear momentum to produce a force, the angular component creates a torque that spins the particles around its axis. A formula for this torque relates to the simple expression $\tau = I\alpha$, where I and α are the moment of inertia and angular acceleration, respectively, for this expression only. The result, based on the absorption of photons with spin angular momentum, is remarkably similar in form to equation (19), i.e.;

$$\boldsymbol{\tau} = \frac{n_{\phi} I_{\omega} \mathbf{u_k}}{2c\varepsilon_0\hbar} \, |\boldsymbol{\mu}^{\mu 0}|^2, \tag{26}$$

where $\mathbf{u_k}$ is the unit vector of $\mathbf{k}$. (However if any significant amount of scattering occurs, in addition to absorption, the result is diminished: retro-reflection of circular polarisation, for example, confers precisely zero angular momentum.) It has to be emphasised that this torque arises only for circularly polarised beams (or, by proportion, for beams of elliptical polarisation); it is the only torque connected to the radiation force of conventional light. For other effects to occur, helically structured beams are required, as detailed below.

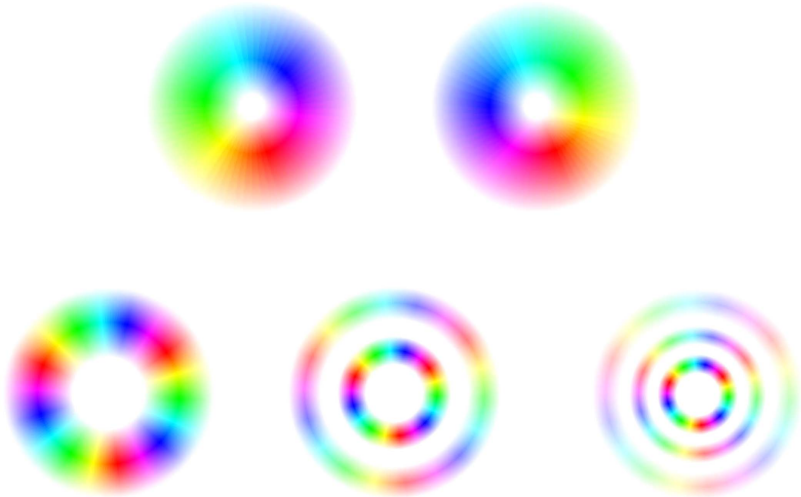

Figure 6. Transverse field distributions for five Laguerre–Gaussian modes, with their associated intensity and phase structure. Top images display $l = 1$ (left) and $l = -1$ (right) with $p = 0$, while the lower images show $l = 3$ with $p = 0$, 1, 2 (left, centre, right). White areas represent zero intensity and the hues denote the phase.

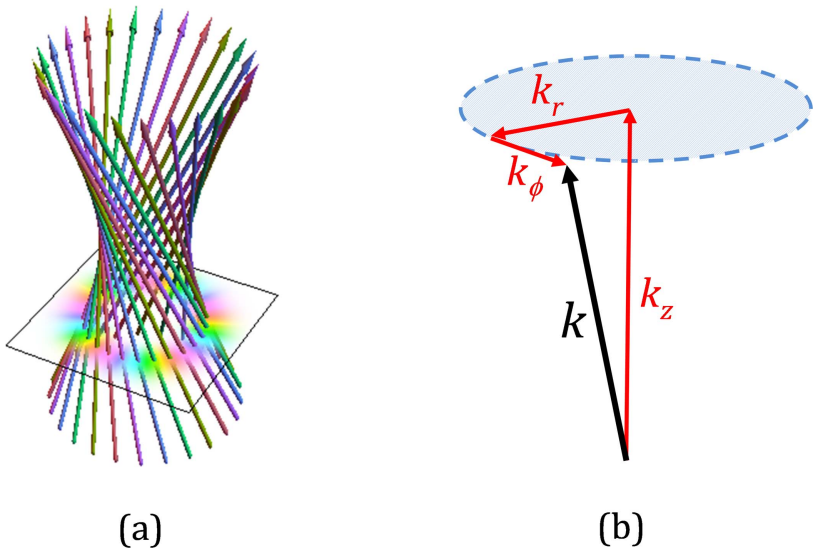

Figure 7. (a) Schematic representation for the focus of an $l = 5$ optical vortex mode. In the transverse plane, hue indicates the phase and brightness the intensity. The bundle of Poynting vectors, depicted as colour-coded arrows, rotates about the beam axis by one fifth of a circle over the space of one wavelength: (b) resolution of a local Poynting vector into axial (z), radial (r) and azimuthal (ϕ) components. Image (a) courtesy of Sonja Franke-Arnold, University of Glasgow and Morgan & Claypool Publishers, © 2016.

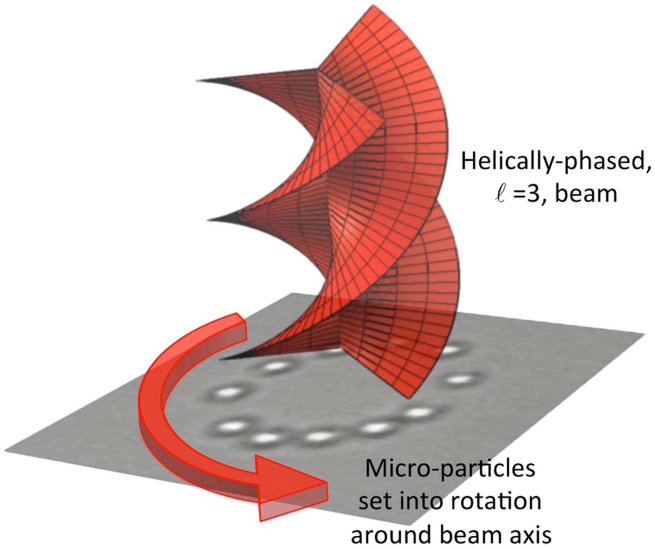

Figure 8. Helically phased laser beam incident on a set of microscopic particles. The latter are trapped within the high-intensity ring of the beam (the centre of an optical vortex beam has zero intensity, unlike Gaussian light) and orbit around the beam axis, as indicated by the red arrow. Image courtesy of Miles Padgett, University of Glasgow and Morgan & Claypool Publishers, © 2016.

4.3. Torque due to orbital angular momentum

When suitably structured, light can also deliver orbital angular momentum, whose quantised form is designated by eigenstates and eigenvalues of the operator $\hat{\mathbf{L}}_{\mathrm{rad}}$. Any such eigenstate is commonly referred to as an *optical vortex*, alternatively called *twisted light*. The key attribute here is that the electromagnetic fields acquire a phase factor of the form $\exp(\pm \mathrm{i} l\phi)$, where ϕ is the azimuthal angle around the beam axis. Generally, the topological charge, l, signifies the number of distinct helical surfaces formed by the optical wavefront, as it winds around this axis within the span of a wavelength. One immediate consequence is that, for $l \neq 0$, there has to be a line of zero intensity along the axis, as a result of an indeterminate phase at the corresponding point in the transverse plane. This feature lends its character to another name associated with this type of beam: singular optics. Light engendered with orbital angular momentum includes Laguerre–Gaussian, and all but the simplest forms of Bessel and Mathieu beams [28–30]; however, in this work, we only examine the former beam type.

Typical examples of transverse field distributions for Laguerre–Gaussian modes are presented in figure 6, which depicts the associated intensity and phase structure. Figure 7(a) gives an impression of how the wave-vector, everywhere pointing along the normal to the wavefront surface, effectively twists around the beam axis. Within the high-intensity rings of such structured light, particles can be trapped and circulate around the central node (figure 8). This orbiting behaviour was theoretically predicted [31] and then experimentally observed [32–36] with an Laguerre–Gaussian beam (which became known as an optical spanner or wrench) [8, 37–39]. The presence of the 'twist' in the optical vortex is the source of the orbital angular momentum that, in turn, produces the torque.

This third type of optical torque is once again most often applied to spherical particles; it can arise due to the radiation force associated with *non-forward* scattering, rather than light absorption. In such a mechanism, in contrast to equation (22), the initial and final system

states are *not* identical, since the created and annihilated photons (with identical energy) travel in different directions. As a result, in terms of the quantum theory, an additional radiation mode is assigned to the system states, so that $|I\rangle = |n, 0; 0\rangle$ and $|F\rangle = |(n-1), 1; 0\rangle$ for the present case. Here, the first and second elements in each state vector denote radiation modes relating to the annihilation and creation events, respectively, and the third element represents the particle state. This subtle change in the initial and final system states, which differs to *forward* Rayleigh scattering, may seem innocuous but it facilitates a physically important differentiation between the radiation and gradient force.

The torque is determined by multiplying the scattering rate (found from the Fermi rule) by the radius of the beam ring, r_b, and the orbital angular momentum per photon $l\hbar\mathbf{u_{kz}}$, where $\mathbf{u_{kz}}$ is the unit vector of $\mathbf{k_z}$; the latter is illustrated in figure 7(b). Following a rotational-average, assuming plane polarised input and summing over all possible polarisations for the scattered light, the following expression is derived for the observed torque;

$$\boldsymbol{\tau} = \frac{Ik^3 l r_b \mathbf{u_{kz}}}{40\pi\varepsilon_0^2 c}(\alpha_{\lambda\lambda}\alpha_{\mu\mu} + 7\alpha_{\lambda\mu}\alpha_{\lambda\mu}). \tag{27}$$

Here, the Greek indices denote the particle frame of reference for the polarisability components (rather than the laboratory frame represented by the earlier Latin indices) and uses the implied summation convention we encountered earlier. This result shows that the optical response of the particle is written in terms of two material parameters, both scalars, in which $\alpha_{\lambda\lambda}\alpha_{\mu\mu}$ represents the square of the trace of the polarisability, tr($\boldsymbol{\alpha}$), and $\alpha_{\lambda\mu}\alpha_{\lambda\mu}$ is a sum of the squares of the nine polarisability components. Larger dielectric particles may be treated with electronic properties closer to those of a bulk material. It is appropriate, in such cases, to engage the linear optical susceptibility rather than the polarisability tensor; the connection between the two is explained in detail elsewhere [40]. In all situations, therefore, there are just two scalar parameters, which together effectively determine the particle's propensity to acquire angular momentum about the vortex beam axis.

5. Discussion

The major aim of this article has been to explain, in a clear manner, the difference between the radiation and gradient forces, and torques, that act on particles when irradiated by laser light. We have presented the different physics involved in each type of force, and the foundation equations that describe them, to assist in this process. Even in contemporary scientific literature, this distinction is sometimes blurred and misinterpreted. Despite these differences, the radiation and gradient forces are completely necessary for the optical manipulation of particles—whether acting alone or in tandem. For example, it has been shown that the radiation and gradient force are vital in atom cooling [41, 42] and optical lattice techniques [43], respectively; while a combination of both are imperative for optical spanners [8]. We have also shown that there are three distinct types of optical torque, of physically different origin. A fuller account of these issues, extending to 'optical binding' effects and including experimental results, is to be found in a forthcoming book [44].

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