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Surface plasmons with phase singularities and their effects on matter

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We explain how a surface plasmon optical vortex can be created when a laser beam with a phase singularity such as Laguerre–Gaussian light is totally internally reflected at the planar surface of a dielectric on which a metallic thin film is deposited. The light field in the vacuum region is evanescent in that it decays with distance perpendicular to the film surface, but significantly, it retains the phase singularity of the original light beam and also the orbital angular momentum carried by that beam. We describe the essential features of such surface optical vortices and discuss how they can, in principle, be used to influence atoms localised in the vicinity of the surface.

1 Introduction A new subfield of optics has recently been established dealing with a novel form of laser light called an optical vortex. Distinguished by phase singularities, such a beam is known to carry a quantised orbital angular momentum $l\hbar$ residing in its azimuthal phase dependence of $e^{il\varphi}$, where l is an integer [1, 2]. This brief report is concerned with optical vortices (typified by Laguerre–Gaussian (LG) beams) generating surface effects which are not only spatially confined, but which are strongly plasmonic as well [3]. The primary aim here is to find the general form of the plasmonic fields and explore their influence on atomic systems near the surface.

In some beautiful recent studies, Tan et al. [4] have demonstrated surface plasmon polaritons generated by optical vortex beams, whilst Gorodetski et al. [5] have established that spin-based plasmonic effects can be created in nanoscale structures by the use of circularly polarized light (due to a geometric phase associated with a spatially varying polarization state). These authors have also shown that a surface spiral surrounding a nanoaperture can confer the necessary phase conditions [6]. The present study aims to determine the behaviour associated with optical angular momentum whose origin is neither photon spin nor surface morphology, the necessary phase properties primarily being conferred by a helical beam structure. Developing earlier work [7], the present focus is on the exploitation of surface plasmonic field enhancement for the control of vicinal atoms. The assumption is that before the LG light is switched on, atoms are trapped vertically, for example by well-known multiple-beam optical methods [8, 9], by a potential well that is larger than kT, and significantly stronger than the potential created by the evanescent light – the latter decaying exponentially in the z-direction. For an atom at rest in the z-direction, the fields conferred by the LG light are essentially two-dimensional, hence motion occurs only in the x-y plane, parallel to the surface.

We begin by outlining the procedure for deriving the plasmonic surface optical fields for the case of a single LG light beam, and the optical forces due to this are identified. We then consider a system of two counter-propagating LG beams in a setup designed to laterally trap adsorbed atoms. Typical parameters are used to determine the optical forces, and to obtain the trajectories for sodium atoms.

2 Evanescent fields Consider first a typical Kretschmann configuration, with a single LG beam impinging from within the dielectric, at an angle of incidence ϕ, on a metallic film of thickness d. From Maxwell's equations the electric field vector components emerge with in-plane polarisation in the three regions of the layered structure, namely the dielectric, occupying the region $z < -d$; the film occupying the region $-d < z < 0$ and the vacuum region occupying the space $z > 0$. These fields are then subject to boundary and phase-matching conditions and the procedure leads to the evanescent field in the vacuum region expressible in cylindrical coordinates in the form

$$\boldsymbol{E}_{klp}(x,y,z>0) = \frac{2A_{klp}(x,y)}{\zeta}\left(\hat{\boldsymbol{x}} + i\frac{k_{\parallel}}{k_{z1}}\hat{\boldsymbol{z}}\right) \times \mathrm{e}^{-i[xn(\omega/c)\sin\phi + k_{z2}d]}\,\mathrm{e}^{-k_{z1}z},$$

where carets denote unit vectors, ω is the frequency of the light, n the refractive index of the dielectric, and

$$\zeta = \left(1 - i\frac{k_{z2}\varepsilon_1}{k_{z1}\varepsilon_2}\right)\cosh(k_{zs}d) + \left(\frac{k_{zs}\varepsilon_1}{k_{z1}\varepsilon_s} - i\frac{k_{z2}\varepsilon_s}{k_{zs}\varepsilon_2}\right)\sinh(k_{zs}d).$$

For a Laguerre–Gausian mode of order l and degree p, the explicit form of the factor A is:

$$\begin{aligned} A_{klp}(x,y) = {} & \frac{\xi_{k00}N_{\ell p}}{\left(2^{p+|\ell|}p!|\ell|!\right)^{1/2}} \\ & \times\left[\frac{\sqrt{2\left(y^2 + (x\cos\phi - d\sin\phi)^2\right)}}{w_0}\right]^{|\ell|} \\ & \times\exp\left[\frac{-(y^2 + (x\cos\phi - d\sin\phi)^2)}{w_0^2}\right] \\ & \times L_p^{|\ell|}\left[\frac{(y^2 + (x\cos\phi - d\sin\phi)^2)}{w_0^2}\right] \\ & \times\exp\left[-i\ell\arctan\left(\frac{y}{x\cos\phi - d\sin\phi}\right)\right]. \end{aligned}$$

In the above, $\boldsymbol{k}_{\parallel}$ is the in-plane wave-vector and k_{z1}, k_{z2}, k_{zs} are propagation wave-vectors in respective regions where the dielectric functions are ε_1, ε_2, ε_s, with regions 1 taken as vacuum and 2 the dielectric of refractive index n, while the metallic film has a frequency-dependent dielectric function in the form $\varepsilon_s = 1 - \omega_p^2/\omega^2$, with ω_p the plasma frequency. The evanescent field is thus specified in full.

3 Effects on atoms We now consider an atom in the vacuum region at position $\boldsymbol{R}(t) = (x(t), y(t), z(t))$, $z > 0$. The electronic properties of the atom are simply cast in the form of a two-level approximation with level frequency separation ω_0 and level width $\hbar\Gamma_0$. The interaction of the atom with the surface plasmon vortex is taken in the dipole approximation, with a Hamiltonian $H_{\text{int}} = -\boldsymbol{\mu}\cdot\boldsymbol{E}_{klp}(\boldsymbol{R})$. The interaction of the atom with the electric field of the plasmonic surface vortex leads to two important dynamical attributes of the motion, namely the Rabi frequency $\Omega_{klp}(\boldsymbol{R}(t))$ and phase $\Theta(\boldsymbol{R}_{\parallel}(t))$. The former emerges in the following form:

$$\begin{aligned} \Omega_{klp}(\boldsymbol{R}) = {} & \frac{2\mu\left(1 + k_{\parallel}^2/k_{z1}^2\right)^{1/2}}{\hbar|\zeta|}\,\frac{\xi_{k00}N_{\ell p}}{\left(2^{p+|\ell|}p!|\ell|!\right)^{1/2}} \\ & \times\left[\frac{\sqrt{2\left(y^2 + (x\cos\phi - d\sin\phi)^2\right)}}{w_0}\right]^{|\ell|} \\ & \times L_p^{|\ell|}\left(\frac{(y^2 + (x\cos\phi - d\sin\phi)^2)}{w_0^2}\right) \\ & \times \mathrm{e}^{-k_{z1}z}\,\mathrm{e}^{-(y^2 + (x\cos\phi - d\sin\phi)^2)/w_0^2}, \end{aligned}$$

while the phase is a function of in-plane variables:

$$\Theta_l(x,y) = \left[\ell\arctan\left(\frac{y}{x\cos\phi - d\sin\phi}\right)\right] - \left(\frac{nx\omega\sin\phi}{c}\right).$$

The Rabi frequency and phase are important for determining the optical forces, which we now consider.

4 Optical forces The total steady state optical force acting on the atom at time t in the vacuum region when its velocity vector is $\boldsymbol{V}(t) = \dot{\boldsymbol{R}}(t)$ is well known from the theory of laser cooling and trapping. It is the sum of a dissipative force and a dipole force, each dependent on both position and velocity:

$$\boldsymbol{F}_{\text{total}}(\boldsymbol{R},\boldsymbol{V}) = \boldsymbol{F}_{\text{diss}}(\boldsymbol{R},\boldsymbol{V}) + \boldsymbol{F}_{\text{dip}}(\boldsymbol{R},\boldsymbol{V})$$

where the mode labels are dropped for convenience. The forces are given by

$$\boldsymbol{F}_{\text{diss}}(\boldsymbol{R},\boldsymbol{V}) = 2\hbar\left\{\frac{\Gamma_0\Omega^2(\boldsymbol{R})\boldsymbol{\nabla}\Theta}{\Delta^2(\boldsymbol{R},\boldsymbol{V}) + 2\Omega^2(\boldsymbol{R}) + \Gamma_0^2}\right\},$$

$$\boldsymbol{F}_{\text{dip}}(\boldsymbol{R},\boldsymbol{V}) = \left\{\frac{-2\hbar\Delta(\boldsymbol{R},\boldsymbol{V})\,\Omega(\boldsymbol{R})\boldsymbol{\nabla}\Omega(\boldsymbol{R})}{\Delta^2(\boldsymbol{R},\boldsymbol{V}) + 2\Omega^2(\boldsymbol{R}) + \Gamma_0^2}\right\}.$$

In the above equations, Δ is the dynamic detuning

$$\Delta(\boldsymbol{R},\boldsymbol{V}) = \Delta_0 - \boldsymbol{V}\cdot\boldsymbol{\nabla}\Theta$$

and $\Delta_0 = \omega - \omega_0$ is the detuning of the light from the atomic transition frequency. The equation of motion of the atom is given by Newton's Second Law, and its solution requires specifying the position and velocity of the atom at $t = 0$, when the beam is switched on. To confine the atom to an angular path in the plane parallel to the surface, we shall need two internally incident beams.

5 Two beams An undesirable feature in the case of a single beam SPOV, as far as interaction with atoms is concerned, is that there is a plane wave with a wave-vector equal to the in-plane component of the incident light traveling along the surface. The corresponding term in the phase function gives a dissipative force, as for a plane wave in free space, but one acting on the atom along the

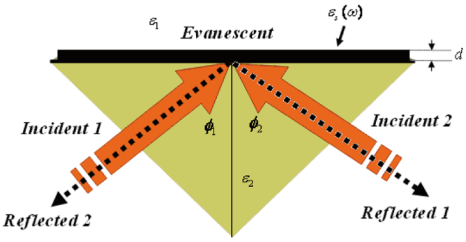

Figure 1 (online colour at: www.pss-rapid.com) Schematic total internal reflection of two LG beams at a planar dielectric interface with a metallic film, creating a surface plasmon from counter-propagating evanescent modes.

x-direction. An arrangement that can eliminate this undesirable motion is a situation whereby the in-plane waves of two beams counter-propagate, as shown in Fig. 1. Here, two LG beams, labeled 1 and 2, are incident at angles $\phi_1 = -\phi_2 = \phi$; both suffer internal reflection at the metallized interface; both have field components within the film, and surface plasmonic components in the vacuum region. We assume that the two beams are identical in modal form, plane of incideence polarization and spatial distribution, differing only in their directions of propagation and the possibility of an associated change in the sign of the angular momentum quantum number *l*.

6 Atom dynamics Having specified the forces acting on the atom centre of mass, we now consider the motion of the atom on the surface. The dynamics again follows a Newtonian equation of motion, driven by a sum of the forces delivered by each beam; the RK4 Runge-Kutta method has been used to numerically solve by iterative convergence the resulting differential equations of atomic motion.

To illustrate the solutions leading to typical trajectories we consider the dynamics of a sodium atom in a surface plasmon, on a thin silver film deposited on glass. The plasmon is formed using two confocal, counter-propagating 'doughnut' Laguerre-Gaussian modes and with parameter values as follows: wavelength $\lambda = 589$ nm; beam-waist $w_0 = 35\lambda$; irradiance $I = 2 \times 10^6$ W m^{-2}; $\varepsilon_1 = 1$; $\varepsilon_2 = 2.298$; $\Gamma_0 = 6.13 \times 10^7$ Hz; $d = 59$ nm; $\mu = 2.6ea_0$ (*e* is the electron charge, a_0 the Bohr radius); $|\Delta_0| = 10^2\,\Gamma_0$. The atom is initially at rest at a position $(x_0, y_0) = (0.5w_0, 0.5w_0)$.

Assuming $l_1 = -l_2 = 1$, $p = 0$ and $\phi_1 = -\phi_2 = 45°$, the calculations show that positive detuning engenders an outwardly spiralling orbital motion leading the atom away from the beam focus not shown). However in the case of negative detuning the trajectory of the sodium atom is very different, as illustrated in Fig. 2. There is now a confinement region in the form of an elliptical concentric valley defined by the intensity distribution, the radial confinement leading to vibrational motion in a radial direction and resulting in an overall zigzag trajectory.

7 Conclusions Our analysis has focused in particular on an evanescent mode generated by laser light with an azimuthal phase dependence and orbital angular momentum – optical features that are typified by Laguerre–Gaussian beams. The fields associated with the surface plasmon mode can be as much as two orders of magnitude stronger than those in the absence of a metal film. In order to ensure the atomic confinement, within the high-intensity regions of the evanescent light, we deploy a second, counter-propagating light beam with the same angular momentum attributes, incident in such a way that the in-plane plane wave effects lead to a trapping of the atom, producing confinement within the high intensity regions of the evanescent fields. The propensity for any such vortices to be concentrated in a small volume, tightly bound to the planar surface, makes such modes interesting – not just in their own right, as essentially two-dimensional modes with rotational features, but also for possible applications. It is our hope that the identification of key features in the anticipated dynamics will spur on experimental activity in this area.

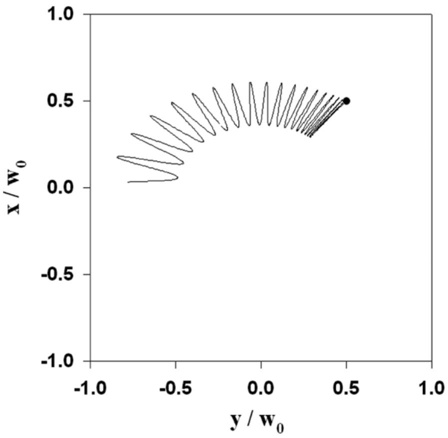

Figure 2 Trajectory of the sodium atom in the evanescent fields generated by counter-propagating LG beams, with negative detuning, at a planar dielectric interface coated with a metallic film. See text for parameter values.

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