Optical vortices produced by diffraction from dislocations in two-dimensional colloidal crystals

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Abstract. A dislocation in a crystal can be described by a Burger’s vector \(b\), whilst a screw dislocation in a laser mode can be described by a topological charge \(l\). By illuminating both optically trapped and self-assembled two-dimensional (2D) colloidal crystals with a Gaussian laser beam, we show a correspondence between crystal and optical dislocations, where the first order diffraction pattern from a crystal with Burgers’ vector \(b = na\) consists of vortex laser modes of topological charge \(l = \pm mn\), \((n = \text{integer}, a = \text{lattice constant and } m = \text{diffraction order})\).

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1. Introduction

Dislocations in crystals play a central role in both the macroscopic and microscopic properties of solids. Understanding the manner in which these dislocations form, change or migrate through a crystal requires a wide range of diagnostic techniques. Attempting to use computer simulations to model dislocations on the atomic scale can be very demanding and is subsequently limited in scope. However, a very promising alternative is the use of microscopic colloidal models [1].

Sufficiently dense solutions of monodisperse hard-sphere colloid will self-assemble into crystals and using optical forces it is possible to produce dislocations in these crystals on demand. By allowing a colloidal suspension to self-assemble into a crystal on top of a patterned substrate, the controlled introduction of dislocations in the crystal is possible (rather than the spontaneous formation of dislocations) [2]. In a similar fashion it is also possible to use an optically defined substrate, an optical potential energy landscape, to force the colloid into an arrangement whereby the lowest energy state is a crystal which necessarily contains dislocations [3]. Additionally it is possible to produce dislocations on demand within a colloidal crystal using indentation [4] or with the help of optical tweezers [5].

Wave dislocations in the form of edge, screw or mixed dislocations can be observed in light in many situations and in fact are ubiquitous in nature [6]. These dislocations are analogous to those found in crystals, exhibiting effects such as the ability to annihilate when pairs of such dislocations meet. Dislocations occur generally in waves including compression waves [6], x-rays [7] and light where knots, loops, braids and links have been observed due to the combination of appropriate optical beams [8]. The dislocations found in light form lines for which the phase is indeterminate, leading to zeros of intensity such as those found in the screw dislocations observed in cylindrically symmetric modes [9]–[11]. Such modes include Laguerre–Gaussian (LG) modes where an integer number of $2\pi$ cycles of azimuthal phase enclose a dark vortex core where the phase is ill-defined [12]. Such laser modes with embedded dislocations may be generated through the use of either direct phase modulation [13] or using a holographic method [14]–[16].

In this paper, we explore the link between dislocations within two-dimensional (2D) colloidal crystals and those embedded within optical fields. We show a correspondence between the Burgers vector of a dislocation in a colloidal array and the topological charge of a diffracted probe laser beam, a link which we then use to investigate a spontaneously formed dislocation in a self-assembled crystal. Additionally we show how, by illuminating a self-assembled colloidal crystal with a reference Gaussian beam, we may locate dislocations within these colloidal crystals and rapidly determine the directions of the slip from inspection of the diffraction pattern. This allows us to determine the direction and magnitude of the Burgers’ vectors of the crystal dislocations. As it is possible to watch the evolution of the topological charge of the optical diffraction pattern in real time, this approach can also be used to investigate the formation and evolution of dislocations in colloidal crystals.

To enable these studies we employ, as a first step, the use of interferometric patterns of light that may align monodisperse colloid into pre-designated arrays using the optical gradient force. Optical trapping of Mie particles, combined with the use of static [17, 18] and dynamic holograms [19, 20], may produce tailored arrays of trapped colloid. It has also been shown that optically trapped particles can be forced into self-assembled crystal geometries via their interaction with the trapping light [17] and that those optically trapped crystals can be used to
diffract a probe laser beam [21]. It has also been shown that it is possible to achieve freezing effects by illuminating a liquid of strongly interacting colloid [22].

The paper is organized as follows: firstly, we describe optical fields with screw dislocations, namely LG light fields in more detail and show how we use them to engineer optical potential energy landscapes that may organize colloidal particles into arrays possessing crystal dislocations. Subsequently we explore the diffraction of a probe laser from these crystals and then progress to observe diffractive light patterns from aggregated colloid (without optical assistance) and make a link between the topological charge or azimuthal phase of the diffracted laser mode and the Burgers vectors of the dislocations within the crystal of interest. We note our study has strong analogies to the determination of the Burger’s vector of an atomic crystal using x-ray diffraction and Moiré techniques [23].

2. LG laser modes

Circularly symmetric LG laser modes form a complete basis set for paraxial light beams. A given mode is usually denoted $LG_l^p$ where $l$ and $p$ are the two integer indices that describe the mode. $l$ refers to the number of $2\pi$ azimuthal phase cycles around the circumference of the mode and $(p + 1)$ indicates the number of radial nodes in the mode profile. These modes have generated much recent interest as, for LG modes with $l \neq 0$, there is an azimuthal phase term $\exp(-il\phi)$ in the mode description that gives rise to a well–defined orbital angular momentum, of $\hbar l$ per photon [24]. This is in addition to any angular momentum the light may possess due to its polarisation state [25]. The field amplitude of an LG laser mode $E(LG_l^p)$ of indices $l$ and $p$ is given by equation (1)

$$E(LG_l^p) \propto \exp \left[ -\frac{ikr^2z}{2(z^2 + z_r^2)} \right] \exp \left[ -\frac{r^2}{\omega^2} \right] \exp \left[ -i(2p + l + 1)\arctan \left( \frac{z}{z_r} \right) \right] \exp \left[ -il\phi \right] (-1)^p$$

$$\times \left( \frac{r\sqrt{\tau}}{\omega} \right)^l L_l^p \left( \frac{2r^2}{\omega^2} \right),$$

(1)

where $z$ is the distance from the beam waist, $z_r$ is the Rayleigh range, $k$ is the wavenumber, $\omega$ is the radius at which the Gaussian term falls to $1/e$ of its on-axis value, $r$ is the radius, $\phi$ is the azimuthal angle and $L_l^p$ is the generalized Laguerre polynomial. The arctan $(z/z_r)$ term describes the Gouy phase of the mode.

LG beams have been used in a wide variety of optical trapping experiments including notably the trapping of low index particles [26] and investigations of the orbital angular momentum of light [27]. LG beams may form novel interference patterns when combined collinearly with a Gaussian beam [28] or another LG beam [29]. These patterns can be rotated such that they can be used to controllably rotate trapped particles [28, 29] as well as to produce simple crystal unit cells [29]. Such structures may be of use for template-assisted crystal nucleation [29, 30].

In our study, we interfere an LG beam and a Gaussian beam at a slight angle such that the subsequent interference pattern consists of a set of linear fringes containing one or more dislocations in the centre of the interferogram. By trapping colloidal spheres in this interferogram using the optical gradient force, we have produced an amplitude modulated hologram equivalent to an LG hologram. This offers a direct and controlled approach to inducing dislocations in colloidal structures, whereby either a dilute solution of monodisperse colloid or a self-assembled
Figure 1. The experimental set-up used to produce the interferogram containing a dislocation. Key; H: hologram, M: mirrors, GP: glass plate, BS: beam splitter, L1-4: lenses, DM: dielectric mirror, A: aspheric lens, F: filter, ×50: microscope objective, 633 nm: HeNe laser, C: condenser assembly, I: incoherent illumination, F: IR filter and CCD: camera.

colloidal crystal is illuminated by the interference pattern between a LG laser-mode and a fundamental Gaussian laser-mode. This produces a set of fringes containing a dislocation such that we can either directly create a miniature crystal dislocation by optically trapping colloid in a dilute sample or force an isolated dislocation, or pair of dislocations, into a self-assembled crystal (akin to that described in [4]).

Illuminating this ‘colloidal hologram’ with a separate low power probe laser, we are able to transform the incident Gaussian mode of the beam into various diffracted orders of LG beams where the azimuthal index of the diffracted LG beam yields direct information about the dislocations embedded within the crystal.

3. Optically trapped structures

The experimental set-up for this arrangement is shown in figure 1. In this set-up, the LG mode \((l = 2)\) produced from a static hologram is interfered, as aforementioned, at an angle with the zeroth-order undiffracted beam from the hologram. This produces a set of linear fringes with a dislocation in the centre. If the LG mode has an azimuthal mode index \(l\), then the magnitude of the Burgers’ vector \(b\) of the dislocation in the fringes, is given by \(b = la\) (where \(a\) is the fringe spacing) [31].
Figure 2. Colloid trapped in the fringe dislocation of an interferogram: (a) shows the strings of the vertical 1D crystal and (b) the liquid order between fringes, (c) and (d) the trapped colloid with superimposed strings (e) the trapped colloid illuminated by the reference beam from the HeNe laser and (f) the resultant diffraction pattern showing vortex beam profiles in the +1 and −1 diffracted orders. By fitting to a line profile of the vortices in (f), the vortices in the first order diffracted beams were found to have a topological charge of $|l| = 2$.

To show that this process is reversible, we explore a trapped array of 5 $\mu$m diameter spheres with a density just below that required to see a self-assembled close-packed crystal (see figure 2). The interference pattern being used to trap the spheres is formed from an LG beam with a topological charge $l = 2$. Hence, the trapped spheres sit in a set of lines with a dislocation in the centre such that there are two more lines at the bottom of the array than the top. Upon illumination
of the trapped-sphere array with a HeNe laser, we can clearly see vortices in the diffracted beams that lie in the same direction as the Burgers’ vector of the crystal dislocation. To find the Burgers’ vector $b$, we must first calculate the lattice constant from the angle of the first order diffracted beams and then multiply this by the topological charge $l$ of the vortex in the diffracted beams.

A full analysis of topological charge $l$ of an LG mode involves interfering the vortex beam with its mirror image $(-l)$ [32]. Alternatively one can use a second analysis hologram that would otherwise produce an LG beam from a Gaussian beam, to obtain information on the mode composition [33]–[35], a technique which has also been used successfully in quantum entanglement experiments [36].

A mode diffracted from an amplitude modulated hologram such as the ‘colloidal holograms’ in this study is a superposition of modes with different radial indices. However, it has been shown that for binary ‘holograms’ of the sort formed by our trapped colloid, the diffracted beam is for all practical purposes equivalent to LG modes with a radial index $|p| = 0$, once propagated into the far-field [14]. In all of the study presented here, the optical diffraction is imaged on to a camera using a microscope objective such that we are always looking at the far-field, allowing us to approximate our diffracted vortex beams as single ringed LG beams. For $p = 0$, a LG laser mode can be described by equation (2)

$$ I(r, z) = \frac{2P_0 e^{-\frac{r^2}{2w(z)^2}}}{\pi |l|!w(z)^{2+2|l|}} (2r^2)^{|l|}, $$

where

$$ w(z) = w_0 \sqrt{1 + \frac{\lambda z}{\pi w_0^2}}. $$

$I(r, z)$ is the intensity dependent on the radial position $r$ and propagation $z$ from a focus. $P_0$ is the total power in a beam of wavelength $\lambda$, $w(z)$ is the beam radius and $w_0$ the beam waist. Hence we find for this case that the horizontal spacing of the trapped colloid is 6.6 $\mu$m and the topological charge $l = 2$ (see figure 3), corresponding to a Burgers’ vector for the trapped sphere dislocation of magnitude 13.2 $\mu$m in the same direction as the diffracted vortex modes. Hence, we see that an edge dislocation of charge 2 in the colloidal array gives us a charge $l = 2$ dislocation in the diffracted beam.

In this case, we have used a hard-sphere colloidal suspension of 5 $\mu$m diameter polystyrene spheres with a size distribution of less than 5%. This leads to close-spacing of the spheres (on the order of a few nanometres), such that we can assume that the diffraction from self-ordering of the monodisperse colloid will correspond to a spacing of $\sim 5 \mu$m (12.4° diffraction angle), allowing us to calibrate the measurements of the diffraction angles from the trapped sphere arrays. It is notable that, due to the lack of long-range interactions, there is not a strong correlation between colloid trapped in adjacent fringes such that the colloidal array forms a crystalline order in the direction orthogonal to the fringes but only has a liquid order parallel to the fringes. However, this demonstration of what could be called a ‘colloidal hologram,’ shows it is possible to use dislocations in a colloidal crystal to obtain dislocations in a diffracted reference beam and that these optical dislocations give us information on the original colloidal dislocation itself.
Figure 3. Determination of the topological charge of the diffracted beam. Here, we see the closest fit which was achieved for a topological charge of \( l = 2 \). It can be seen that there is only a very slight contribution to the shape of the mode from the superposition of LG modes with radial indices \(|p| > 0\), which lead to the small secondary peaks either side of the main structure of the beam.

Notably in the above case the reference Gaussian beam is aligned such that the dislocation is approximately centred on the beam axis. This results in an optical vortex that resides close to the centre of the diffracted beam. If however the reference beam is not centred on the dislocation, then we see the dark spot shift away from the centre of the diffracted beam, giving an off-axis optical vortex [37, 38].

4. Dislocations in self-assembled colloidal crystals

The dislocations in self-assembled colloidal crystals were either those formed spontaneously in a monodisperse sample or, when the density of dislocations was low, by the introduction of impurities (3 \( \mu \)m diameter spheres as an impurity in a suspension of 5 \( \mu \)m diameter spheres at a relative concentration of 1/15 or less.) Introducing impurities locally disrupts the nearest neighbour spacing such that we get a lowest energy state where impurities are located at dislocations.

Figure 4 shows the results obtained when a dense monodisperse solution of 5 \( \mu \)m diameter polymer spheres were allowed to crystallize into a close-packed 2D arrangement (NB—we have used the full four Miller indices for describing these crystals to allow for the out of plane illumination of the crystal to be described with the same index system). Figures 3(a)–(c) map three separate planes of the crystal, allowing us to make comparisons with what we see in the diffraction pattern in figure 3(e). The dislocation is clearly seen once the planes are mapped out. The dislocation extend out to the nearest grain boundaries in the crystal. We can see that the diffracted orders in the [01 \( \bar{1} \) 0] and [10 \( \bar{1} \) 0] directions corresponding to the [01 \( \bar{1} \) 0] and [10 \( \bar{1} \) 0]
Figure 4. A self-assembled crystal of hard-sphere colloid. (a)–(d) show the [01 \bar{T}0], [1 \bar{T}00], [10 \bar{T}0] and [1 \bar{3} 20] strings respectively. By illuminating the crystal with a reference Gaussian beam (e) along the [0001] direction it is possible to locate the dislocations and rapidly determine the directions of the slip by looking for vortices in the diffracted orders as seen in panel (f), which allow us to determine the direction and magnitude of the Burgers’ vector of the dislocation.

planes contain vortices but the [1100] plane, which is parallel to the Burger’s vector, leads to diffraction into Gaussian spots containing no vortex. So, by studying the diffraction pattern from the principal crystal planes we are able to determine the size and direction of the dislocation (the Burgers’ vector).

The topological charge of the vortex (found using the technique described in section 2) multiplied by the lattice spacing, calculated from the diffraction angle, tells us the magnitude of the Burgers’ vector of the dislocation. For the case shown in figure 4, we find that the spacing between the [10 \bar{T}0] set of crystal planes is 4.3 \mu m and the topological charge of the vortices are |l| = 1, giving a Burgers’ vector of magnitude 4.3 \mu m. The direction of the burgers vector is the [10 \bar{T}0] crystal direction.

We also observe l = 1 vortices in the first order diffracted beams from the more closely spaced [1210] and the [12\bar{3}0] sets of planes. This shows how we may use optical dislocations to obtain information about crystal dislocations in a quick and simple manner. Close inspection of figure 4(c) reveals that in some diffracted orders the vortex is shifted off-axis as a result of a slight misalignment of the dislocation and reference beam centres. As the beam reference beam is moved around the vortices also shift within the diffracted orders, eventually aligning to the centre of a diffracted beam when the reference beam centres on the dislocation. This effect could be used to find the exact location of dislocations. Though it is not possible to identify clearly in figure 4, we would also expect that higher order diffracted beams would contain a higher topological charge [31]. (The second order diffracted beam will have twice the charge of the first order, etc.)
5. Conclusions

We have explored the generation of screw dislocations or vortices within a probe laser beam diffracted through a colloidal crystal with dislocations. We used optically trapped arrays as well as self-assembled crystals of hard sphere colloid and in both cases were able to find the Burgers’ vector of a dislocation through a combination of the diffraction angle and the topological charge of the vortices in the first order diffraction pattern.

This link between the charge of the optical vortex and the magnitude of the Burgers’ vector of the crystal has allowed us to demonstrate a new and powerful optical method for the detection and analysis of dislocations in colloidal crystals, one that could be used in studies such as those on the formation and evolution of dislocations in colloidal crystals. This method may be used to study the formation and evolution of dislocations in colloidal crystals [2, 4]. The technique is analogous to Moiré techniques already used in x-ray diffraction studies of atomic crystals [23], further extending the toolkit with which colloidal models can be used to study crystal properties.

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