# Hollow Bessel-like beam as an optical guide for a stream of microscopic particles

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**Abstract:** Current aerosol sample injection methods for coherent x-ray morphology suffer from excessive sample consumption due to the dispersion of the aerosol. To remedy this we propose here a high aspect ratio optical funnel by using a hollow Bessel-like beam with variable divergence, which may reduce sample consumption significantly. We present estimated optical forces exerted on the particles in the transverse plane, depending on various experimental conditions. We show that light pressure imposed by a funnel formed with 4.2 W continuous wave laser is sufficient to divert a stream of 2 μm polystyrene particles travelling ~50 m/s by ~$1.5 \times 10^{-3}$ rad.



**OCIS codes:** (140.3300) Laser beam shaping; (140.7010) Laser trapping; (140.2600) Free-electron lasers (FELs); (350.4855) Optical tweezers or optical manipulation.

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## Introduction

Structural biology is in urgent need of high-throughput methods for structure determination of proteins, particularly those that are large, complex, and difficult to crystallise. X-ray free-electron laser (FEL) based serial femtosecond crystallography is an emerging method with potential to rapidly advance challenging fields such as membrane protein structural biology [1,2]. Due to the extremely short x-ray pulse duration, FELs provide the potential to overcome the effects of radiation damage, which is frequently the principle resolution-limiting factor. The atomic-scale imaging afforded by x-ray scattering provides a 'seeing is believing' route that powerfully addresses questions in disciplines ranging from physics and chemistry through materials science to geophysics and structural biology.

Coherent x-ray radiation of the current generation of FELs reaches into the soft x-ray regime at 4.16 nm in the FLASH facility at the Deutsches Elektronen-Synchrotron (DESY) in Hamburg, Germany, and the Linac Coherent Light Source (LCLS) in Stanford, USA, presently operates up to 120pm (first harmonic), offering unprecedented capabilities for structural characterization of radiation-sensitive bioparticles, including ultrafast and irreversible dynamics [1–4]. Intended applications in near future include the imaging of complex, single biological molecules with ~10 fs time resolution.

Coherent diffractive x-ray imaging of individual biological macromolecules with FELs requires a "touch-free", high-precision particle injection method to deliver individual biological molecules or nanoparticles to the micrometer-sized focus of the x-ray beam. This method must typically operate in medium- to low-vacuum regimes (> $10^{-5}$ mbar), Currently existing aerodynamic lens systems produce single-particle diffraction by streaming particles across the interaction region, which are stochastically intercepted by the x-ray beam, see Fig. 1. In experiments performed thus far, the average imaging efficiency (fraction of particles intercepted by the X-ray beam) is $<10^{-7}$ on average. For samples that cannot be produced in high abundance, simple aerodynamic lens systems render FEL single-particle experiments infeasible, since the determination of a high-resolution three-dimensional structure will likely require > $10^6$ individual diffraction patterns. Improved methods should aim to provide precise delivery of biological macromolecules and droplets of sub-micron dimension, preferably as a single-file, mono-disperse stream that is phase-locked to an external timing signal.

To address this challenge we are developing an optical Bessel-like beam for use as an "optical pipeline", with a variable-diameter hollow core and an axial-to-lateral aspect ratio up to ~2000, that can be used to guide particles with a spatial precision of less than a few µm over centimetre-long distances. We show ways to control the beam divergence aiming to focus the stream of particles by radiation pressure, analyse the forces acting on the particle in the beam and present preliminary results on diverting a stream of particles by a counter-propagating hollow Bessel beam collinearly aligned with the axis of the stream.

## Guiding particles with optically induced forces

There were recently a number of publications demonstrating various techniques of guiding aerosol particles in air [5–9]. Two complementing forces act on particles optically trapped in a gaseous environment. In addition to the intuitive force applied by momentum exchange with photons (radiation pressure), thermal effects via photophoretic force can also play a major role [10–13]. Both mechanisms of particle guiding in our experimental conditions are discussed in more detail below.

### *Radiation pressure*

The concept of guiding particles with an optical pipeline in vacuum is based on the macroscopic force applied due to radiation pressure $P_{rad} = (1+R)I/c$, where $R$ is the reflection coefficient, $I$ is the laser intensity, and $c$ is the speed of light. This implies that under radiation pressure the particles tend to move along the intensity gradient to areas of lower irradiation. For simplicity, and at the same time for evaluation of the minimum amount of optical force, we further assume highly absorbing particles, $R = 0$. Any reflection from the surface increases the radiation pressure up to the pressure on totally reflecting surface $2I/c$.

The force $F_{rad}$ exerted by the beam on a particle is $F_{rad} = P_{rad}S$ where $S$ is the projected surface area of the particle. The effect of polarisation of light on transport and levitation of spherical particles for the cases of linear, circular and spatially variant (azimuthally or radially oriented) polarisation states were considered in detail using Fresnel formulas in [14,15]. Typical magnitudes of the radiation pressure forces used to manipulate and guide micron size

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particles with a 1-W vortex beam range from femtonewtons to piconewtons, depending on the beam-to-particle size ratio.

*Photophoretic force*

An additional thermal force emerges in a gaseous media due to a possible non-uniform temperature distribution within the irradiated particle. Surrounding gas molecules interacting with the 'hot' side of the particles acquire more energy and higher speed than those reflected from the colder side; as a result the particle gains a net momentum directed towards lower irradiation intensity. This force, named photophoretic force [10,11], depends on the particle size relative to the mean-free path of gas molecules (characterised by a Knudsen number *Kn*), optical properties, and the thermal conductivity of the particle.

Photophoresis converts light to mechanical energy via a chain of events: (i) – Absorbed light heats part of a particle's surface inducing a temperature gradient across the particle, (ii) – the molecules of a surrounding gas bounce off the heated part with increased kinetic energy, and (iii) – the resulting imbalance in momentum exchange with the environment propels the particle away from regions of high light intensity. Simple estimates show that in experiments of guiding micro-size particles with a hollow-core laser beam [16], even moderate light absorption can produce photophoretic force several orders of magnitude larger than the radiation–pressure force [12,13,17].

Here we present experimental results conducted with micron-size particles injected into the vacuum chamber with an aerodynamic lens stack [18,19]. The air pressure in the aerodynamic lens gradually reduced from atmospheric down to ~$10^{-3}$ mbar. Photophoretic force induced by a laser beam is about one to two orders of magnitude higher than the light pressure at atmospheric pressure. It increases further with reduced pressure by about another order of magnitude, up to the level where $Kn = 1$ [11], before it starts going down. For the particle size of ~2 µm $Kn = 1$ is reached at about 10 mbar. At the output nozzle of the aerodynamic lens the pressure is of the order of $10^{-3}$ mbar, where the photophoretic force is expected to be of the same order of magnitude as the force of light pressure.

Both radiation pressure and photophoretic forces are linearly dependent on light intensity, and both forces are collinear to the intensity gradient pushing particles in the direction towards minimum intensity. This is opposite to the optical field/polarizability gradient force that is weak under the experimental conditions employed here [20,21]. For this reason the most appropriate shape of the beam compressing a stream of particles is an optical 'funnel' with a variable angle of divergence. Optimally the beam intensity and geometry – the size and the angle of divergence, could be precisely adjusted to impose the force required to compress a stream of particles with particular particle size, speed of flow, and the jet dimensions. Below we present the results of our first attempts to construct such an optical funnel to compress a jet of particles directed to the focus of the x-ray FEL laser.

**Formation of an optical funnel with controlled divergence**

We applied the concept of the optical pipeline to design an appropriate beam to compress the stream of particles ejected by an aerodynamic lens into the vacuum chamber in the FEL experiments. These experiments impose a number of limitations on the way the optical guiding can be constructed. First, the laser beam should counter-propagate the jet of particles with its axis precisely aligned to the axis of the particle jet and with micron-scale precision passing the focal spot of the FEL x-ray beam. Second, the beam should have a controlled divergence to be able to funnel the particle jet coming out of the aerodynamic lens and to guide them into the x-ray focus. The jet diameter is of the order 50 µm – 100 µm, ideally it should be compressed to less than 1µm (the size of the x-ray focal spot at LCLS presently is 200 nm). Third, the large scattering angles of x-rays captured in coherent diffractive imaging implies that the distance from the aerodynamic lens nozzle to the x-ray focus should be no smaller than 10-20 mm. And finally, the intensity of the beam should be high enough to

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provide optical forces of sufficient strength to quickly direct the particles exiting the aerodynamic lens nozzle at speeds of the order of 50 m/s – 100 m/s to the desired focal point.

Summing up, the experimental conditions of optical compression of a jet of particles in FEL experiments imply that the doughnut-shaped funnel beam should have a waist of a few µm, should expand to ~50-100 µm over a distance of 20 mm (the length-to-diameter aspect ratio ~1,000, beam divergence 3.5 mrad), and that the intensity of the beam should divert the one-micron particles moving with the speed of ~50-100 m/s to the x-ray focus of the FEL – see Fig. 1. The doughnut-shaped profile of a first order Bessel-like beam seems to be the most plausible option to satisfy all these requirements simultaneously.

The ideal Bessel beam derives from a class of exact solutions to the free scalar wave equation, expressed in the form of zero-order Bessel functions of the first kind, $J_0$ [22,23]. The beam shape has a central core with a series of concentric rings. A salient feature of the Bessel beams is that the intensity profile of the central core is independent of propagation distance $z$.

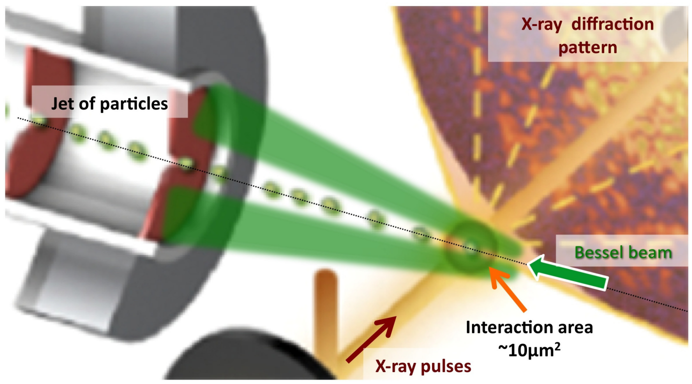


Fig. 1. Conceptual scheme illustrating the compression of a particle stream injected into the interaction chamber with an aerodynamic lens, using a counter-propagating first-order Bessel beam – the 'funnel'. The background image in this figure is adapted from Ref [19].

Finite apertures of optical elements form Bessel beams with finite dimensions. The closest experimental approximation to the ideal Bessel beams, the Bessel-like or Bessel-Gauss beams have the central non-diffracting maximum over a limited propagation distance. The intensity along the central axis gradually decreases with distance. The maximum range for a focused Bessel-like beam can be approximated [22,23] as $z_{\max}^{\text{Bessel}} \cong \pi d_A d_f / 2\lambda$, where $d_f = 2\lambda/\pi \text{NA}$ is the beam diameter at the focus $f$ of a lens with a numerical aperture $\text{NA} \cong nD/2f$, $n$ is the refractive index of the media, $\lambda$ is the wavelength and $d_A$ is the lens aperture. This maximum range of propagation is much larger than the Rayleigh length $z_R^{\text{Gauss}} = \pi w_0^2/\lambda$ of more conventional Gaussian beams, as $d_A \gg d_f$.

Building structural beams with optical elements requires high laser intensity, which is critical for applications using optical forces at the sub-piconewton range. The experimental setup is shown in Fig. 2. A first-order Bessel beam was constructed by illuminating an axicon with a vortex beam with topological charge $l = 1$ formed by a phase plate. We generated a slowly diverging Bessel-like beam with diameter ranging from 5 µm to 50 µm over a propagation distance of 20 mm, i.e. the divergence angle is ~$10^{-3}$ rad. The beams were analysed with an imaging system consisting of a de-magnifying telescope and a CCD camera.

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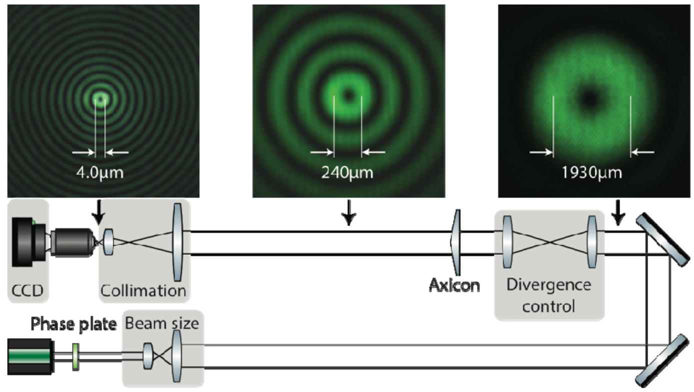


Fig. 2. The experimental setup generating and characterizing a first-order Bessel beam with a combination of a spiral phase plate and an axicon in one arm.

A vortex beam with a single topological charge was formed by propagating the expanded linearly polarized beam of a 532 nm continuous wave laser (Coherent Verdi G5SLM) operated at 100 mW through a 16-step phase plate (Holo/Or Ltd). A Galilean telescope constructed from a pair of 150 mm lenses was used to control the divergence of the beam entering the axicon with a base angle 0.5°: increasing the incident beam divergence caused an increase in the divergence of the output beam. The beam was then demagnified with a telescope comprised of a lens (f = 600 mm) and a microscope objective (WD = 10.6 mm). The imaging system consisted of a 20x microscope objective and a CCD camera placed on a translation stage. It was designed to characterize the resulting field after the last telescope in three dimensions. The dependence of the central core diameter on the distance from the axicon and the resulted beam divergence is shown in Fig. 3.

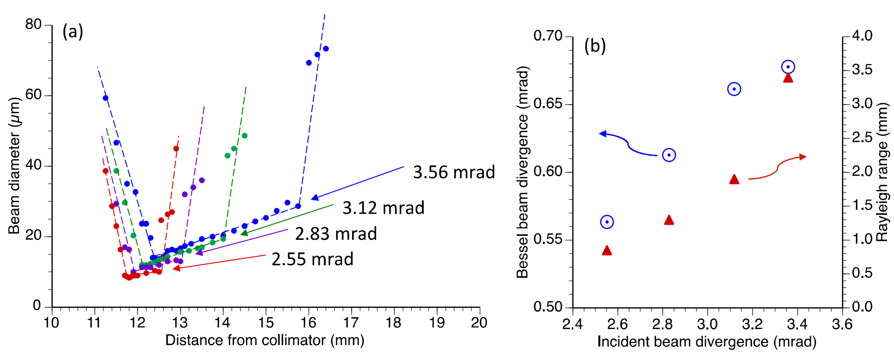


Fig. 3. a) – Dependence of beam diameter on the propagating distance for the beams generated by a combination of a spiral phase plate with an axicon at different divergence of the entrance beams (shown in the graph); b) – dependence of the Bessel beam divergence (circles) and the Rayleigh length (triangles) on the divergence of the incoming beam.

The increase in divergence of the central core of the beam resulted in corresponding shortening of the Rayleigh range of the beam, which is clearly seen in Fig. 3. To obtain the desired beam size, the magnification of the last telescope has to be large, which results in a short Rayleigh range of the demagnified Bessel beam. With the desired beam size and divergence, the propagation distance of the beam is even shorter, typically less than 0.5 mm, which is impractical for our applications. For this reason, it seems that the best option is to form the required Bessel-like beam by using a computer controlled SLM.

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## Experiments on diverting a jet of particles

We performed preliminary experiments aiming to divert a jet of particles exiting the aerodynamic lens into a vacuum chamber with a counter-propagating Bessel-like beam collinearly aligned against the jet. We suspect that the overall forces will have contributions from both radiation pressure and photophoretic effects when the beam is aligned with the particle jet. However, using a counter propagating guiding Bessel-like beam with the particle jet, we can first investigate the effects of radiation pressure of the particles via an opposing particle. The pressure can be measured by position of particle displaced from the jet. The experiments were performed with 2-μm polystyrene spheres continuously injected with an estimated speed of 50 m/s – 100 m/s and collected onto a microscope slide covered by a sticky gel located at a distance of 66 mm from the nozzle. The jet cross-section was estimated by measuring the particle distribution on a gel under a microscope as ~70 ± 5 μm in diameter. A 4-W CW Bessel beam with diameter of ~75 μm was formed by a helical phase plate and an axicon with a base angle of 0.5°, directed opposite to the jet propagation through the microscope slide and aligned collinearly with the jet. In this configuration the injected particles enter the central ring of the Bessel beam.

By shifting the jet in the transverse plane by 25 μm, 50 μm, 75 μm and 100 μm relative to the Bessel beam we were able to repeatedly and reliably shift the position of the particles arriving on the slide by up to 80 μm when the laser beam was "ON" and return the jet to the original path when the laser was "OFF" – see Fig. 4 and Media 1. However, in these preliminary experiments we did not observe the effect of 'focusing' of the particle jet. One of the reasons was possible misalignment of the laser and particle beams, which may occur when the chamber is pumped on (alignments were carried out at atmosphere, whereas the chamber must be under vacuum during aerosol injection).

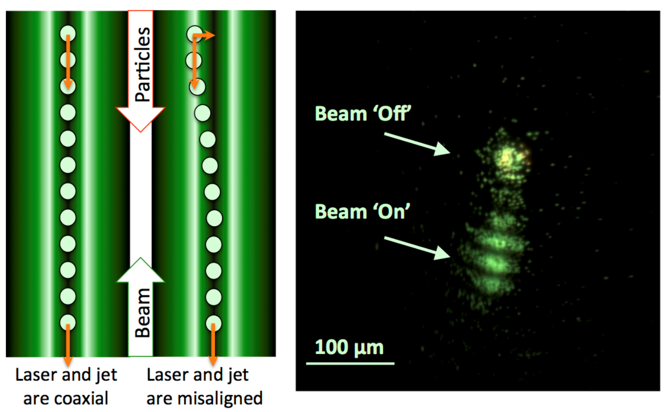


Fig. 4. The concept for the beam diverting experiments with a 4.2 W coaxial Bessel beam counter-propagating the jet of particles is shown on the left for the cases of coaxial and misaligned beam to the particle jet. The arrows show the direction of particle movement in the beam. On the right is an image of the particles deposited on a sticky gel-pack microscope slide for better visualization. The laser beam is misaligned by 75 μm transversally, relative to the jet axis. The top spot was deposited on the slide while the beam was in the 'off' state. The lower spot corresponds to the beam turned 'on', demonstrating the shift of particle path by radiation pressure imposed by the beam. The image shows the beam in 'off' position. The 'on' spot has a strip-like structure because it is illuminated by the rings of the Bessel beam in 'off' state with a minimal power needed only for illuminating the deposited particles (Media 1).

From the measured shift of the beam of 80 μm and reasonably assuming a particle velocity of 50 m/s moving over 20 mm we can roughly estimate the light pressure imposed by the beam on a 2-μm particle (1.05 g/cm$^3$). A back-of-the envelope estimates taking the energy required to move the particle by the light intensity, yields ~1 pN force necessary to deflect the jet from the original path. However, more accurate measurements of the particle speed, beam

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alignment, and particle size are required to predict accurately the particle path and the required laser beam parameters. In a simple approximation, particle trajectories could be calculated by estimating radiation pressure exerted by the guiding beam. As the size of the guided molecules approaches the trapping wavelength, further calculations necessitate the use of Mie scattering, T-matrix or similar methods. In particular, in situ diagnostics of the particle velocity distribution in the jet measured with a fast camera is needed, which is out of scope of this paper and will be presented elsewhere.

Nevertheless, this proof-of-principle demonstration shows a hollow-core Bessel beam from a continuous wave laser source does apply sufficient radiation pressure to divert the particle jet over relevant length scales for FEL experiments.

## Conclusions and discussions

We constructed a highly-collimated optical micro-pipeline, or laser funnel, from a hollow-core Bessel-like beam with waist diameter ~4 μm and aspect ratio up to 1000 with controlled level of divergence, aiming to guide micron-size particles to a precise position in the micro-focus of x-ray free electron laser. The control over the divergence of the Bessel beam was performed by varying the beam divergence.

We measured laser power in a series of cross-sections of the beam relative to the incident power, and reconstructed an intensity map of the beam, which will be further used to calculate the optically guided particles trajectories in this beam. The experimental tests were conducted with 2 μm size polystyrene spherical particles to demonstrate, as a proof-of-principal, the ability to change the particles trajectories under the optical pressure and to evaluate the optical force. The estimated optical forces are of the order of ~1 pN, this corresponds to a laser intensity on the particle surface of ~$10^5$ W/cm$^2$.

There are several important problems to be resolved before the micro-pipeline developed can be applied as a reliable means to compress the jet of particles by at least an order of magnitude and to significantly improve the precision of the particle injection. First, it would be advantageous to have better control over the intensity distribution in the funnel, aiming to increase the intensity at the entrance of the jet into the optical funnel. Second, calculations of particle trajectories guided by the pipeline depend on a number of unknown parameters such as the particles optical properties, size distributions, particle velocity and the flow rate from the aerodynamic lens. In particular, in situ diagnostics of the particle path in the funnel still has to be developed. In addition, knowledge of local gas pressure at the exit of the aerodynamic lens (where the gas freely expands) and thermal properties of the particles would allow evaluating the contribution of the thermal photophoretic force. Furthermore, other potentially valuable architectures for optical trap should also be considered and studied. Fast (relative to particle speed) scanning of a zero-order Bessel beam with engineered intensity [24] would mimic a constant trapping potential matching particular particle properties. Another possible option is constructing a series of concentric optical vortices.

Summing up, rapid development of SLMs with improved efficiency and speed offers a wide range of new opportunities to optically guide micro- and nanoparticles with unprecedented precision in vacuum and in gaseous media. We expect that the hollow-core optical beams will significantly, by orders of magnitude, reduce the particle consumption in X-ray morphology experiments with free-electron lasers.

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