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## Report Title

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# Trapping of individual airborne absorbing particles using a counterflow nozzle and photophoretic trap for continuous sampling and analysis

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We describe an integrated opto-aerodynamic system and demonstrate that it enables us to trap absorbing airborne micron-size particles from air, hold them and then release them, and to repeat this sequence many times as would be appropriate for continuous sampling of particles from air. The key parts of the system are a conical photophoretic optical trap and a counter-flow coaxial-double-nozzle that concentrates and then slows particles for trapping. This technology should be useful for on-line applications that require monitoring (by single particle analyses) of a series of successively arriving particles (e.g., from the atmosphere or pharmaceutical or other production facilities) where the total sampling time may last from minutes to days, but where each particle must be held for a short time for measurements (e.g., Raman scattering).  [http://dx.doi.org/10.1063/1.4869105]

There is a need for improved on-line instruments for studying and monitoring aerosols. Aerosols affect the earth's radiation budget and global climate by scattering and absorbing light and modifying clouds and precipitation (by acting as cloud and ice nuclei). Some aerosols transmit diseases of human, animals, and plants. Some are allergenic or toxic. For measurements of atmospheric aerosols and other complex mixtures of particles, there is often a strong benefit in measuring particles one at a time in order to observe and enumerate the various aerosol types, including minority species. Although spectra (mass, fluorescence, and laser-induced-breakdown) have been measured for individual particles flowing through an air sampler, Raman spectroscopy can provide far more information than fluorescence spectra, and supply information complementary to mass spectrometry. However, Raman emission is very weak. Collecting an adequate Raman spectrum from a single micron-sized particle requires a combination of high NA collection optics and measurement times of at least seconds. These requirements necessitate confining the particle to a very small volume during the measurement. Single-particle Raman spectra have been measured from individual particles sampled from the atmosphere and held in an electro-dynamic trap.[1] However, motion of the trapped particles and other problems made the measurements difficult, and we know of no reports of improved measurements in the intervening 16 yr.

Various methods for optical levitation, trapping, or/and manipulation of micron-sized particles and nanoparticles in water or air have become powerful tools in aerosol science, chemistry, physics, and biology.[2–16] Gradient forces of a tightly focused laser beam (laser tweezers) can trap a dielectric particle in water[3] or air.[4] Optical trapping of a microparticle is typically more difficult in air than in water.[14] The buoyancy of water helps balance the gravitational force on particles. Particles in water tend to remain suspended longer than those in air and have much smaller settling velocities. Even in a closed chamber without intentionally generated air currents, it is still difficult to optically trap a moving microparticle in air. In addition, the scattering force on a microsphere from a laser beam is much higher in air than in water because of the higher relative refractive index of the microparticles in air, so a large numerical aperture microscopic objective (e.g., N.A. = 0.85 or 0.95 (Ref. 15)) is required to generate a stronger gradient force than scattering force for stable trapping. Photophoretic forces can be orders of magnitude larger than radiation pressure forces on an absorbing particle[6–8] and can be used to levitate and stably trap absorbing particles in air.[6–8,16] Aerosol particles made of carbon nano-foam or agglomerates,[7,8] nigrosin, Johnson grass smut spores, riboflavin, and carbon black[15] have been captured and stably trapped in air. The trapping volume can be formed as a low-light-intensity biconical region totally enclosed by the high-intensity light at the surface of the bicone. Particles within the bicone are trapped by photophoretic forces pushing them toward the low-intensity center of this region.[7,8,16]

Although there is a clear need for optical technologies to continuously sample particles from air by trapping them for sufficient times to measure their Raman spectra or observe time evolution phenomena, no such sampling and trapping system has been demonstrated. Optical trapping techniques used to study the physical, chemical, or biological properties of one or a few representative particles in air generally capture and trap the particles from a large group of particles (e.g., a few to 1000's). The particles to be trapped typically must have similar properties and are initially placed on a substrate or in a container and then forced into the air in a short time[14] to generate very high particle concentrations for trapping. Such low efficiency, passive particle trapping approaches are not adequate for continuously sampling and trapping. For many on-line applications (e.g., atmospheric

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aerosol monitoring), particles with different properties must be analyzed from a series of successively arriving particles. To achieve a sufficient particle sampling rate, existing on-line analytical systems (e.g., single-particle fluorescence spectrometers and mass spectrometers) generally concentrate and focus the aerosol into a localized jet which flows into an interrogation region where the particles are analyzed one-by-one as they rapidly flow through. Using a nozzle to concentrate and focus aerosol particles usually requires high velocities (many times larger than the settling velocity in still air). Directly trapping such fast-moving particles from air would require far more intense laser beams than those used in typical laboratory trapping studies, and, would be far more difficult than in typical trapping studies if it is even possible. Therefore, to develop a useful on-line analytical system based on trapping particles from air, the key technology needed may be to overcome the difficulties in trapping caused by the problematic airflows needed to concentrate particles and focus them into a jet for delivery into the trapping region.

This Letter demonstrates a technology that enables us to concentrate particles from air, focus the air that carries them into a narrow jet, slow the jet and particles in it, trap one or more absorbing particles from a region where the jet velocity has become very small, hold the particle for as long as needed, and release the particles to be ready to trap the next one. Once such a system is automated, it should be able to continuously sample successively arriving aerosol particles and hold them for measurements. It combines two key elements: (1) A hollow cone surrounded by a conical high-intensity light surface that can trap absorbing particles carried into the cone by an air stream. (2) Two co-axial nozzles operated in a counter flow mode so that: (i) particles are aerodynamically focused and concentrated into a small-diameter jet and (ii) particles are slowed (by a counter flow of air which slows the jet in which they are entrained) until their vertical velocity approaches zero near the vertex of the hollow optical cone, so that particles move slowly enough to be trapped by photophoretic forces that are able to balance the small drag and gravitational forces.

Figure 1 illustrates the experimental arrangement. The trapping light source is a continuous-wave argon ion laser at 488 nm (Lexel Laser, 95-SHG). It produces a linearly polarized Gaussian beam. Its output power can be adjusted by changing the plasma tube current and/or the neutral density filters. The laser is separated into two beams (50:50) by a non-polarized beam-splitter. One beam is formed into a conical trapping region surrounded by a high-intensity-light at the surface of the cone. The other beam is formed into a laser sheet to cover the cone as a high-intensity light wall on the open side of the hollow cone. The purpose of the sheet is to prevent other absorbing particles from entering the trapping region and hold the trapped particle. The laser beam for forming the cone is cleaned and re-collimated using a 2-mm-diameter diaphragm, two lenses (f = 25 mm and 180 mm), and a 500-$\mu$m-diameter pinhole. The expanded ($\sim$15 mm in diameter) collimated beam is transformed into a hollow beam (doughnut-shaped transverse intensity profile) by passing it through two axicons separated by $\sim$50 cm (Del Mar Photonics, cone angle 175°). The re-collimated hollow beam

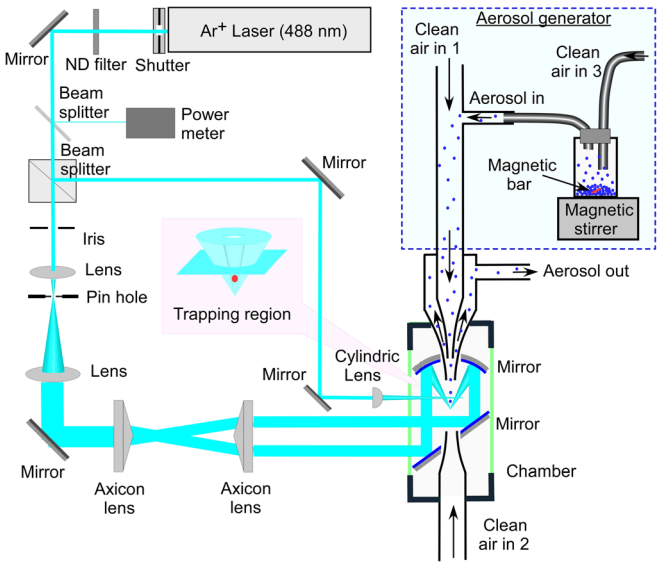


FIG. 1. Schematic of the experimental setup for continuously sampling, trapping, and releasing airborne micron-size particles for on-line observation or measurement. The photophoretic trapping region is formed by a hollow conical beam. Once a particle is trapped, the trapping region can be covered by a laser sheet to prevent additional particles from entering the trap and holding the trapped particle by photophoretic force (see inset in the middle). Particles are aerodynamically focused and concentrated into a localized jet by the inner nozzle and slowed by a counter flow of air surrounding the jet from the outer nozzle and moves particles into the trapping region very slowly.

is $\sim$20 mm in diameter with a low-light-intensity center ($\sim$7 mm in diameter). Fig. 1 shows the 2-dimensional cross section of the laser beam along its axis, so the hollow beam is indicated by two broad lines. The hollow beam, introduced through a quartz window into an air-tight chamber, is reflected by an elliptical mirror at 45°, propagates up to a concave spherical mirror (f = 19 mm, diameter=25.4 mm), and reflects from it to form the low-light-intensity conical region surrounded by the high-intensity-light at the surface of the cone. The high-intensity-light is focused to a spot approximately 5 mm below the tip of the outer nozzle (counter-flow nozzle). The inset in the middle of Fig. 1 labeled "trapping region" illustrates this region in 3D. Each mirror inside the chamber has a center hole to let the aerosol or air flows pass through. The four sides of the chamber are covered by quartz windows to provide a large solid angle (>0.3 $\pi$ sr for each side) for observation as shown in Fig. 2(b).

Figure 2(a) illustrates the trajectories of particles after they exit the outer nozzle. Figure 2(a) shows a 5 s exposure image (recorded with a CCD) of approximately 50 tryptophan particles (8-$\mu$m average diameter) illuminated by a light sheet from a pulsed 527-nm laser operating at 1 KHz. Most of the particles are focused into an aerosol jet which has 220-$\mu$m diameter at approximately 1.5 mm below the outer nozzle tip. Inset (i) in Fig. 2(a) shows the image of a 100-$\mu$m-diameter fiber for rough scale. Inset (ii) in Fig. 2(a) shows a typical trajectory of a moving particle. The bright spots are from the light scattered by the particle at the position when it is illuminated by the pulsed laser. Each millisecond (ms) the particle moves from one spot to the next. Therefore, the speed of the moving particle can be estimated to be the distance between two adjacent spots divided by

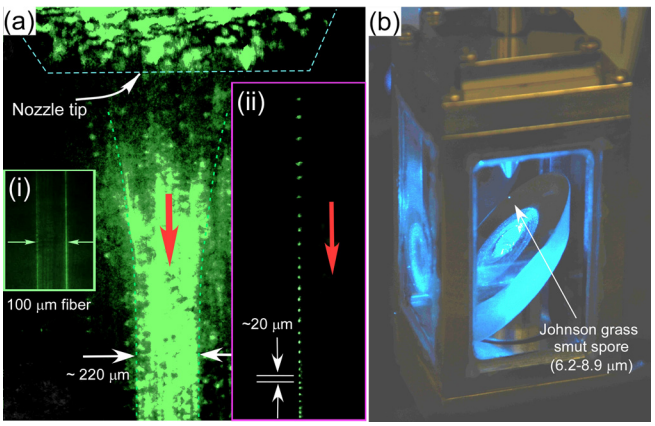


FIG. 2. (a) 5 s exposure image from the moving trajectories of about 50 tryptophan particles (average diameter 8 $\mu$m) recorded by a CCD. Particles are visualized by the illumination of a pulsed 527-nm laser sheet at 1 KHz. Inset (i) is the image of a 100-$\mu$m-diameter vertically oriented fiber for a scale factor. Inset (ii) is the trajectory of a typical particle moving within 1.5 mm below the outer-nozzle tip. It is slowed from 80 mm/s near the top to 20 mm/s in less than 1.5 mm. (b) A single Johnson grass smut spore (6.2–8.9 $\mu$m in diameter) trapped stably in air.

1 ms. In this illustration, the particle slowed from about 80 mm/s to 20 mm/s in approximately 23 ms and in less than 1.5 mm. It illustrates how rapidly the particles are slowed. Typically in concentrating and focusing particles for air sampling the air velocities are greater than 1 m/s, often 20 times greater. If a micron-sized particle needed to be trapped in an airflow of 1 m/s, the drag force on the particle would exceed achievable cw optical forces on the particle. Drag would make it extremely difficult, if not impossible, to optically trap particles moving at these speeds. Here, the airflow is slowed by introducing an outer coaxial-nozzle and pulling the air in a reverse direction from that typically used focusing nozzles. As the air speed decreases, the particles are slowed by the drag forces resulting from the particle's inertia tending to remain constant, i.e., to keep the particle velocity greater than that of the airflow.

Eventually, the downward component of the air velocity and the particle velocity decreases to zero and changes sign. The particles and the air then move laterally out of the jet and into the counter flowing air stream which is drawn back up into the nozzle, and they exit through the outer nozzle. The trapping region is set a little above the center of the zero-velocity point. This design reduces the downward Stokes drag force on the particle in the region where the particle is trapped so that a photophoretic force is strong enough to stop the particles and trap them. The small drag from the downward airflow along the central line, the gravitational force and optical (mostly photophoretic) forces push the particle to the point where it is trapped. Particularly, photophoretic forces push particles toward the lowest light-intensity region, which is the central line of the aerosol jet, and the net photophoretic force acts on the particle in the opposite direction of gravitational forces; while optical pressure forces push particles towards the highest light-intensity point, which is the vertex of the laser beam, along the laser propagation direction. For absorbing particles, optical pressure forces are typically small compared to the photophoretic forces, so the main forces that work together to trap an absorbing particle are photophoretic force, drag force, gravitational force, and optical pressure force here. The combination of the designs (optical trapping cone and counter-flow coaxial-double-nozzle) makes it possible to readily capture and trap particles continuously drawn into the chamber. Fig. 2(b) shows a single Johnson grass smut spore captured and trapped stably in air within the chamber. For highly absorbing particles, the photophoretic force and drag force (from the very weak downward airflow immediately above the region where the downward flow stops) are the main forces for trapping. For absorbing particles with much weaker absorptivity, radiation pressure forces may also contribute significantly to the trapping forces, pulling the particle toward regions with high optical intensity gradients near the vertex of the cone. We have found non-absorbing particles to be very difficult to be captured and trapped with the present experimental conditions.

Figure 3 (Media 1) shows how 8-$\mu$m diameter tryptophan particles move one by one inside a 1 cm $\times$ 1 cm quartz cell. In this case, there is no optical trapping cone. These particles were also visualized by illuminating with the pulsed 527-nm laser operating at 1 KHz. The particles exit the tip of the outer nozzle at approximately 100 mm/s, which was already slowed from approximately 2 m/s exiting the inner nozzle. Typically, a particle moves rapidly until it approaches the turning point about 5 mm below the nozzle. It then moves upwards first slowly and then more quickly. The particles exit the chamber through the outer part of the outer nozzle. This movie shows that super micron particles could be focused into a localized small-diameter jet with a velocity that goes to zero. Such a design provides a technique that can concentrate and focus particles into a localized aerosol jet and then deliver them into a small region at a very low speed (the vertical component of its velocity goes transiently to zero). This coaxial-double-nozzle running in a counter flow mode should be also useful for trapping particles using electrodynamic or ultrasonic forces (rather than optical forces) for continuous sampling.

Figure 4 (Media 2) shows how the system can repeatedly capture a particle from a jet of micron-size aerosol particles that are continuously drawn into the chamber, and then trap the particle and release it (by blocking and unblocking the trapping laser beam). The trapping laser power is 50 mW. The particles shown are aggregates (approximately 20-$\mu$m average diameter with large variance)

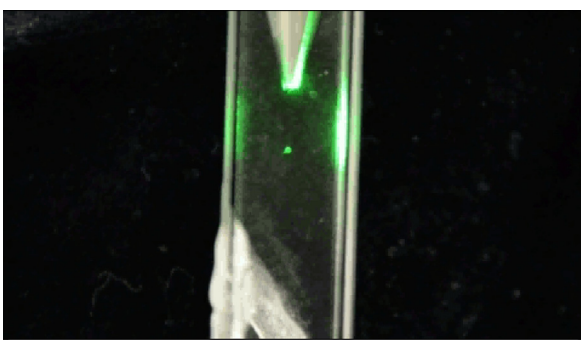

FIG. 3. (Media 1) 8 $\mu$m diameter tryptophan particles move one by one under the control of the counter-flow coaxial-double-nozzle. Particles were visualized by the illumination of a pulsed 527 nm laser operating at 1 KHz (Multimedia view) [URL: http://dx.doi.org/10.1063/1.4869105.1].

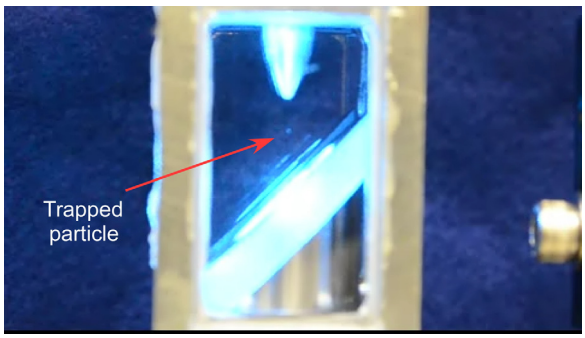


FIG. 4. (Media 2) Aggregates (~20 $\mu$m in diameter around) of multi-walled carbon nanotubes are introduced into the optical chamber. Different individual particles are captured, trapped, and released (by blocking and unblocking the trapping laser beam) at 50 mW laser power (Multimedia view) [URL: http://dx.doi.org/10.1063/1.4869105.2].

of multi-walled carbon nanotubes (MWCNTs) which have an outside diameter less than 7 nm, an inside diameter approximately 2–5 nm, and a length approximately 10–30 $\mu$m. During a 1 min time period, more than 10 particles have been captured, trapped, and released. As seen in Fig. 4 (Media 2), a new particle can be captured and trapped from a jet within 1 s, once the laser is unblocked. The trapping rate could be increased by reducing the time that the particle is in trap and the time the laser is blocked. The trapped particles appear as bright or dim spots (with stronger or weaker scattering intensities), which may be from larger or smaller particles, or from a particle scattering more or less as it is trapped a little further or closer to the highest-intensity light of the vertex. As seen in the movie, when the cover lid (the laser sheet beam) is not on, sometime the image from a trapped particle became brighter and brighter, probably because additional nanotube particles agglomerated with the trapped particle, and the overall super-micron particle kept growing. Occasionally, a trapped particle was knocked away by a newly arriving particle, which either got trapped or moved away from the trapping region. Sometimes the trapped particle has a small-amplitude oscillation and moves within a distance far smaller than the observed particle dimension. The trapped particle eventually reached a stable balance with less or no observable oscillation. It could become very still when the airflow was turned off, and/or when the cover laser sheet "lid" was on to keep the coming particles out. We also found that Johnson grass smut spores (6.2–8.9 $\mu$m in diameter) could trapped and released with a high trapping rate.

We have used the technology described here to repeatedly sample, trap, and release light-absorbing airborne micron-sized aerosol particles that are continuously drawn into a chamber. This technology has the potential to be developed into a new analytical instrument for on-line observation or measurement.

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[1]R. Vehring, C. L. Aardahl, G. Schweiger, and E. J. Davis, J. Aerosol Sci. **29**, 1045 (1998).
[2]A. Ashkin, Phys. Rev. Lett. **24**, 156 (1970).
[3]A. Ashkin and J. M. Dziedzic, Appl. Phys. Lett. **19**, 283 (1971).
[4]A. Ashkin, J. M. Dziedzic, J. E. Bjorkholm, and S. Chu, Opt. Lett. **11**, 288 (1986).
[5]R. Omori, T. Kobayashi, and A. Suzuki, Opt. Lett. **22**, 816 (1997).
[6]M. Lewittes, S. Arnold, and G. Oster, Appl. Phys. Lett. **40**, 455 (1982).
[7]V. G. Shvedov, A. S. Desyatnikov, A. V. Rode, W. Krolikowski, and Y. S. Kivshar, Opt. Express **17**, 5743 (2009).
[8]A. S. Desyatnikov, V. Shvedov, A. Rode, W. Krolikowski, and Y. S. Kivshar, Opt. Express **17**, 8201 (2009).
[9]S. Chu, J. E. Bjorkholm, A. Ashkin, and A. Cable, Phys. Rev. Lett. **57**, 314 (1986).
[10]A. Ashkin and J. M. Dziedzic, Science **235**, 1517 (1987).
[11]G.-Chavez, D. McGloin, H. Melville, W. Sibbett, and K. Dholakia, Nature **419**(6903), 145 (2002).
[12]J. B. Wills, K. J. Knox, and J. P. Reid, Chem. Phys. Lett. **481**, 153 (2009).
[13]L. B. Kong, P. F. Zhang, G. W. Wang, P. Setlow, and Y. Q. Li, Nat. Protoc. **6**, 625 (2011).
[14]K. C. Neuman and S. M. Block, Rev. Sci. Instrum. **75**, 2787 (2004).
[15]T. Li, Fundamental Tests of Physics with Optically Trapped Microspheres (Springer Science, NY, 2013), pp. 21–28.
[16]Y. L. Pan, S. C. Hill, and M. Coleman, Opt. Express **20**(5), 5325 (2012).