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The optical trapping of airborne hydrocarbon droplets from an oil mist

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This paper describes the optical trapping of hydrocarbon droplets in air using a high numerical aperture objective lens (NA = 1.4) to tightly focus a continuous wave Nd:YAG laser ($\lambda = 1064$ nm). The trapping efficiency ($Q = 0.42$) was sufficiently robust to enable capture of droplets directly from the turbulent flow of an oil mist generated using a commercial ultrasonic nebulizer. Droplets could then be manipulated in three dimensions for periods in excess of 30 minutes. Ultrasonic nebulizers are commonly used for respiratory therapy; hence the captured oil droplets were in the respirable size range of 1–7 μm. To enable preliminary studies of droplet interactions two optical traps were created using acousto-optic deflection techniques to multiplex the laser beam. These traps were independently manipulated to force a collision that resulted in rapid droplet coalescence where the larger droplet was retained in the optical trap. Information on the chemical nature of the trapped droplets was also acquired by changing the laser wavelength to 514.5 nm and collecting the Raman spectra.

Keywords: Optical trapping; Aerosol droplet; Levitation; Laser tweezers; Raman tweezers; Ultrasonic nebulization

1. Introduction

The optical levitation of liquid droplets in the gas phase was pioneered by Ashkin and Dziedzic [1–7] and the methodology has been used to study a number of low-volatility liquids [3, 8–11], such as glycerol and dioctyl phthalate. The combination of optical levitation with Raman spectroscopy [8, 12, 13] has provided a powerful tool for *in situ* chemical analysis of these aerosol particles. To date, these techniques have been used to study time-resolved compositional analysis in polymerization [14, 15], acid–base [16] and ozonolysis reactions [17]. Rapid and accurate determination of the droplet size has been of particular interest. The size of optically levitated aerosol droplets can be

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determined through fluctuations in the levitating radiation pressure [14], changes in polarization ratio of the laser beam [9], and through structural and morphology dependent resonance effects in the Raman spectra of optically trapped spheres [10, 11, 18–22]. The sizing techniques have been utilized in further studies into absorption, growth, evaporation, and temperature changes [23] on isolated aerosol droplets.

There are two common strategies employed to optically levitate droplets. The first method balances gravitational forces with those of the radiation pressure from an upward pointing and weakly focused laser beam. The numerical aperture (NA) of the focusing lens is typically [24] in the range of 0.01–0.25 and thus the convergence angle, θ, of the focused laser light is between 0.5° and 12.5°. Under these conditions the average reported axial trapping efficiency, Q, is 0.05 [24–26]. With this efficiency, great care has to be taken to present single droplets to the levitation trap to prevent collisions from other particles or disruption from air currents. These techniques use vibrating orifices or ink-jet printer heads to produce chains of single droplets. The reported trapped droplet diameters are in the size range of 12–150 μm (diameter), although sizes of a few microns have been recorded [9]. The vertical position of the trapped particle is dependent upon laser power, but this can be stabilized using a feedback control between a position sensitive diode and the laser power control [14, 27].

The second strategy for optical levitation uses high numerical aperture microscope objective lenses, which tightly focus the laser beam to form a single beam optical gradient trap [27–29]. Optical trapping using this method has been documented for a number of years, but is normally associated with capturing particles dispersed in liquid media. Importantly, gradient trapping should not depend on balancing gravitational forces and can be achieved in any optical orientation. Magome *et al.* [24] have used a NA 1.3 ($\theta = 60°$), oil-immersion, ×100, microscope objective lens to capture water droplets nucleating *in situ*. These researchers observed the trapping of an aerosol droplet at the focal point of the laser beam with a significant increase in trapping efficiency as Q is reported to be 0.46. A similar optical technique is used here to create a trapping environment that utilizes the greater trapping strengths to capture single droplets from a mist passing the point of focus but where no special preparation of the aerosol sample is required. Our development enables practical utilization of the trapping techniques to capture aerosols found in the environment for application to areas such as climate change, pollution studies and combustion science. The chemical processes in these applications can then be studied in real time through acquisition of Raman spectra. The technique has already been used in trapping environmentally relevant aerosol droplets [17, 22] and here we report the technical details used during the original construction of the apparatus.

2. Experimental method

The optical trap was created by focusing a continuous wave, TEM_{00}, Nd:YAG diode laser (Laser 2000) with a wavelength of 1064 nm and a variable power output of up to 1 W. From the laser source the beam was expanded and directed into a Leica DM IRB microscope where it was reflected upwards, using a dichroic mirror, such that the back aperture of the objective lens was slightly overfilled by the laser beam.

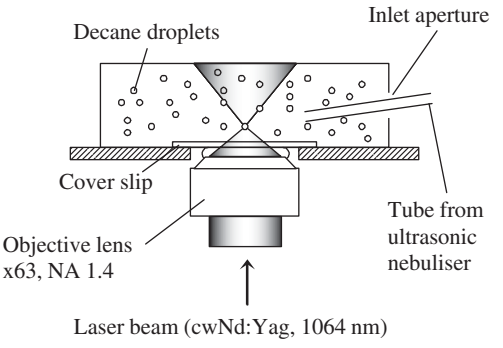


Figure 1. Schematic diagram of the optical trapping arrangement and sample chamber.

The lens used was an oil-immersion objective with NA = 1.40 and $\theta = 67^\circ$ working distance. The beam could be steered and multiplexed in the plane of focus using a pair of orthogonally mounted acousto-optic deflectors (Isle Optics Ltd). An oil mist (n-decane, refractive index = 1.41) containing droplets in the size range of 1–7 μm was generated using a commercial hand-held ultrasonic nebulizer (Omron NE-U07). A schematic diagram of the chamber and trapping environment is shown in figure 1. Experiments were performed at a temperature of $19 \pm 0.5^\circ$C. Images of the droplets were taken from below the droplets using a CCD camera mounted on to the conventional optics for inverted microscopy. The magnification was calibrated with a stage graticule to enable sizing of the trapped droplets. The alterations to the apparatus for acquiring Raman spectra are fully detailed in previous work [22, 30]. Raman spectra were collected for 10 seconds using a trapping power of 13 mW.

3. Results and discussion

Injection of an oil mist into the sample chamber resulted in the observation of numerous droplets passing the field of view. Within a few seconds most of these droplets had collided with the upper surface of the cover slip to form a thin condensed film. The thickness of the decane film could be determined from the reflection of a focused laser spot from the glass–decane and decane–air interfaces. The decane film varied in thickness from 5 to 15 μm, depending on the volume of mist injected into the sample chamber. The position of the focal spot was adjusted to be above the decane film (see figure 1).

After the decane film had formed, a second injection of the oil mist resulted in advection of droplets across the laser focal region until a single airborne droplet became trapped at the point of focus. After the initial trapping event there was a short period of droplet growth through droplet–droplet collisions. Observation of the collisions showed that they did not result in ejection of the droplet from the trap. Droplet growth slowed as the droplet concentration diminished and the mist dissipated. The maximum diameter attained through collision was 10 microns. The droplet could be held and manipulated for periods in excess of 30 minutes. Throughout this period,

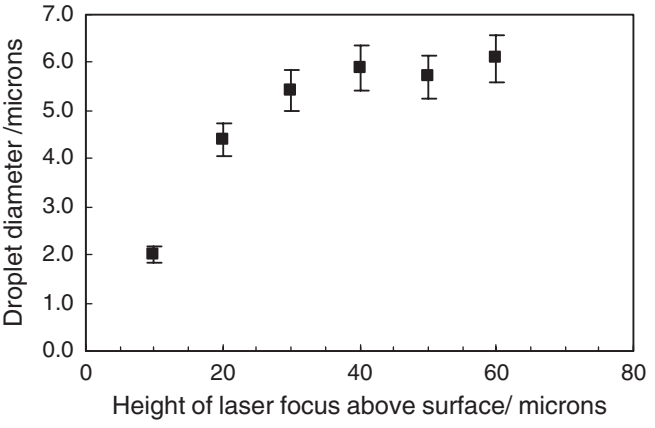
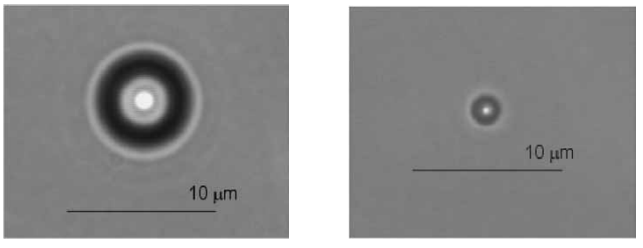


Figure 2. Diameter of the optically trapped aerosol droplets as a function of the height of laser focus above the cover slip.



(a) (b)

Figure 3. Images of trapped aerosol droplets of (a) 6.7 μm and (b) 2.0 μm diameter.

droplet volume remained constant at the laser trapping power of 20 mW indicating that evaporation through laser heating was minimal [22]. The droplet volume was determined from analysing the positions of the droplet edges from the video picture which were accurate to the nearest pixel. The magnification of the microscope, as determined by measuring the spacing of a stage graticule, corresponded to 1 pixel being equivalent to 76 nm. Therefore, as the droplet size remained unchanged (to the nearest pixel) over a period of 30 minutes the size change was less than 0.1 nm s^{-1}. To assess the robustness of the optical trapping, the chamber cover was removed and the droplet exposed to a turbulent flow across the sample area, even under these conditions, the droplet remained optically trapped.

The final size of the trapped droplet was found to be dependent on the position of the laser focal point above the cover slip. Five droplets were trapped at each height, the image recorded and the average size of droplet determined. The variation in the droplet sizes at equilibrium is shown in figure 2 and images of the droplets are displayed in figure 3. The error bars in figure 2 correspond to the standard deviation of size variation. Droplet size appears to be dependent on the relative position of the

focal point of the laser beam to the decane film surface. Smaller droplets were trapped close to the surface. The larger diameter droplets were unable to be trapped immediately above the decane film surface without undergoing coalescence with bulk decane.

The trapping efficiency was determined by gradually reducing laser power whilst holding a series of droplets with an average of 5.9 μm (diameter). The average power just sufficient to retain the droplet was 0.5 mW. The droplet did not change significantly in either size or focal position during the reduction of laser power. The axial trapping efficiency, Q, is [24]

$$Q = \frac{fc}{nP}$$

where f, c, n and P are the trapping force, velocity of light, refractive index of medium, and laser power, respectively. For levitation, the trapping force along the vertical axis is

$$f = mg = \frac{4}{3}\pi r^3 \rho g$$

where m, r, ρ and g are the droplet's mass, radius and density and the gravitational constant, respectively. This results in the value of Q as 0.42 which is higher than that of conventional levitation traps and close to that reported for previous studies using larger droplets.

Having successfully trapped the airborne droplets the beam steering and manipulation capability of the tweezers apparatus was then used to study droplet interactions. Here, two optical traps were simultaneously created using a pair of acousto-optic deflectors to multiplex a single laser beam [31]. These traps could be independently steered and were used to capture, manipulate and control the approach of two droplets as shown in figure 4. Coalescence was observed and recorded using video capture techniques. Image analysis of the resulting frames indicated that the total droplet volume remained constant during coalescence. This observation has been verified

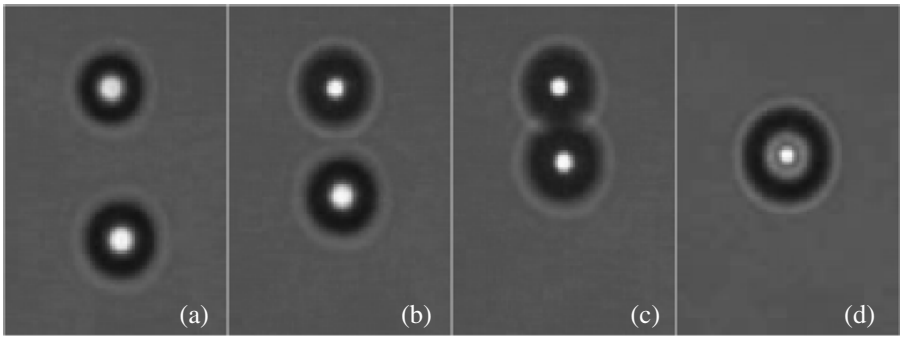


Figure 4. Image sequence showing (a), the capture of two aerosol droplets that are then brought together (b) and (c), until the droplets coalesce (d).

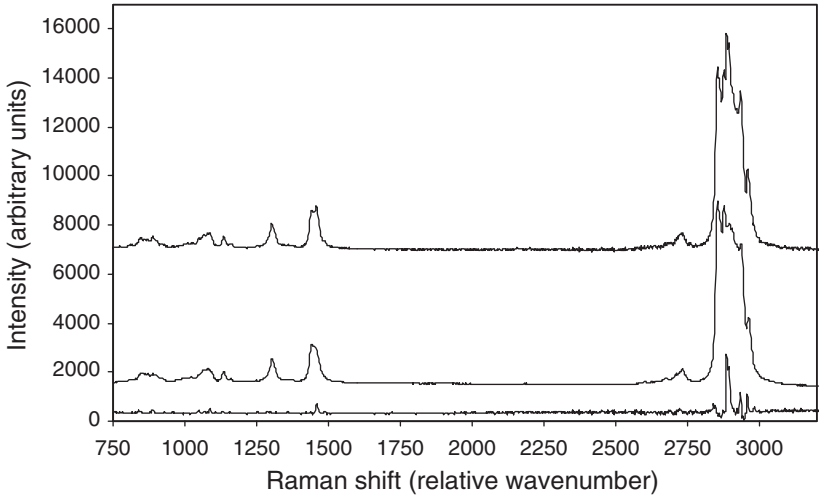


Figure 5. Raman spectra of an optically trapped droplet (top) in comparison to bulk decane (middle) with the difference spectrum (bottom) showing morphology dependent resonances at 1459, 2845, 2886, 2934 and 2958 rel cm^{-1}.

by related studies using identical apparatus [22]. The coalescence process was noted to occur over a time-scale of less than one video frame (less than 40 milliseconds) and future work will aim to establish the precise rates of coalescence.

The Raman spectrum acquired from the airborne decane droplet is shown in figure 5 in comparison to bulk decane. For bulk decane the peak positions are in agreement with those reported as reference values. However, with trapped decane droplets a number of peaks in the C–H stretching region (2800 to 3000 rel cm^{-1}) appear to be very sharp and were noted to shift on each acquisition. On closer inspection of the difference spectrum (bulk decane subtracted from the decane droplet) these resonances are also seen to extend to lower wavenumber. Such a phenomenon has been previously reported [22] with airborne water droplets where they are attributed to morphology dependent resonances within the optically trapped droplets.

4. Conclusions

Within this paper we have demonstrated a method for the robust optical trapping of single and multiple aerosol droplets from a nebulized mist of droplets, having sizes commensurate with respirable therapy applications. This has been achieved using a high numerical aperture microscope objective lens. We have shown that the trapped droplet sizes can be controlled by either adjustment of the depth of the laser focus in the sample chamber or by manipulation and coalescence of multiple droplets. Chemistry within the droplet species can also be probed using Raman spectroscopy. The combination of these capabilities has resulted in a flexible

and robust technique for the study of aerosol droplets in a range of environmental applications.

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