# Optical sculpture: controlled deformation of emulsion droplets with ultralow interfacial tensions using optical tweezers

Andrew D. Ward,*[a] Mark G. Berry,[a] Christopher D. Mellor[b] and Colin D. Bain*[c]

*Received (in Cambridge, UK) 14th July 2006, Accepted 31st August 2006*
*First published as an Advance Article on the web 28th September 2006*
**DOI: 10.1039/b610060k**

**We report a technique for deforming micron-sized emulsion droplets that have ultralow interfacial tensions, by the manipulation of multiple optical trapping sites within the droplets.**

Laser-based optical trapping has become a widespread technique for manipulation and control of colloidal and biological particles.[1] The advantages of the technique lie in the precise and non-destructive control of microscopic particles without the requirement for sample perturbation by mechanical means. Optical trapping has been primarily used to measure forces exerted externally on a trapped object. Here we describe the deformation of a single emulsion droplet using multiple optical tweezers to provide a means for applying internal forces.

In general, small objects trapped with optical tweezers do not deform because the radiation pressure of the continuous wave (CW) lasers used in optical trapping is feeble compared to the Young's modulus of most solids or the Laplace pressure within a micron-sized liquid droplet. Thus, droplets form and retain their perfect spherical geometry to minimise the surface area, and hence the Gibbs free energy, for a given volume. Deformation studies have been performed on liposomes and red blood cells,[2] in which a thin lipid bilayer membrane encapsulates the cell. Distortions in the membrane are then possible because of the low values of the elastic shear and bending moduli of the bilayer.

It has been known, however, since the experiments of Ashkin and Dziedzic 30 years ago[3] that focused (and especially pulsed) laser beams can cause measurable deformations in a planar or pseudo-planar (*i.e.* large radius of curvature) surface. Recently, there has been a flourish of interest in deforming surfaces this way. For instance, Zhang and Chang[4] studied the effect of pulsed lasers on macrodroplets of water, Casner and Delville[5] showed that giant deformations could be induced with continuous wave lasers if the interfacial tension was low, and Mitani and Sakai[6] developed laser interface manipulation as a means of measuring interfacial tensions.

In this paper we describe how large deformations can be achieved in droplets of an oil-in-water emulsion with a low-power CW laser. For this phenomenon to occur the interfacial tension at the oil–water surface has to be reduced to a value comparable with the force constant of the optical trap, which is typically $10^{-5}$ to $10^{-6}$ N m$^{-1}$. Since oil–water interfacial tensions are around 0.05 N m$^{-1}$ for an alkane, the interfacial tension has to be reduced by four orders of magnitude. Such ultralow tensions are achieved by adsorption of surfactants under conditions where the emulsion is close to the microemulsion phase boundary (where the interfacial tension vanishes). The system we chose comprises AOT–hexane–brine. Temperature was used to fine-tune the interfacial tension and large deformations were observed with a laser power of $10^{-2}$ W.

The optical trapping experiments were carried out in the Laser Microscopy Laboratory of the Central Laser Facility at the Rutherford Appleton Laboratory, Oxfordshire, UK. For these initial experiments we prepared an oil-in-water emulsion by blending together heptane, water, salt (0.05 M NaCl) and the surfactant Aerosol OT (1 mM AOT). Heptane (Sigma-Aldrich, >99%) was used after passing through an alumina column three times and AOT (Sigma-Aldrich, >99%) was used as received. The oil was added to the surfactant solution and the two briefly agitated to begin emulsification. At this point, the emulsion is in a dynamic state where the droplets are a few microns in diameter and the heptane has not yet microemulsified. The interfacial tension of this emulsion system has been characterised by Aveyard[7] using spinning drop tensiometry. It was also used by Mitani to demonstrate laser induced deformation of a planar oil–water interface.[6] A region of ultralow interfacial tension ($<3 \times 10^{-6}$ N m$^{-1}$) is known to exist between 20 °C and 23 °C. In our study, the emulsion was injected into a 100 μm deep flow cell as shown in Fig. 1. Single droplets were captured near the upper surface as droplets in the emulsion started to cream. The trapped droplet was dragged vertically down into the bulk solution to be isolated from other droplets. All deformation experiments were performed at 20.0 ± 0.5 °C.

The experimental apparatus has been reported previously.[8] In brief, a 1 W Nd:YAG laser (Laser 2000) at 1064 nm was passed through a pair of perpendicular acousto-optic deflectors (AOD), whose function was to steer the laser beam in the *x*–*y* plane. The first-order diffracted beam was expanded and then directed into an inverted microscope (Leica, DM-IRB) *via* a dichroic mirror and microscope objective (Leica, ×63 water immersion, NA = 1.2). The signals applied to the AOD were multiplexed using computer software to generate up to four optical traps with individually controllable trapping position. Laser powers at the focus were 11–27 mW; at these values laser heating is expected to be minimal (<1 °C).[9] Dividing the optical traps into multiple positions is assumed to lower the average laser power at each position in the same ratio. The images were recorded using a CCD camera and

---

[a] *Lasers for Science Facility, CCLRC, Rutherford Appleton Laboratory, Chilton, Didcot, UK OX11 0QX. E-mail: a.d.ward@rl.ac.uk; Fax: +44 (0)1235 445693; Tel: +44 (0) 1235 446751*
[b] *National Institute for Medical Research, Mill Hill, London, UK NW7 1AA*
[c] *Department of Chemistry, Durham University, South Road, Durham, UK DH1 3LE. E-mail: c.d.bain@durham.ac.uk; Fax: +44 (0)191 3342051; Tel: +44 (0) 191 3342138*

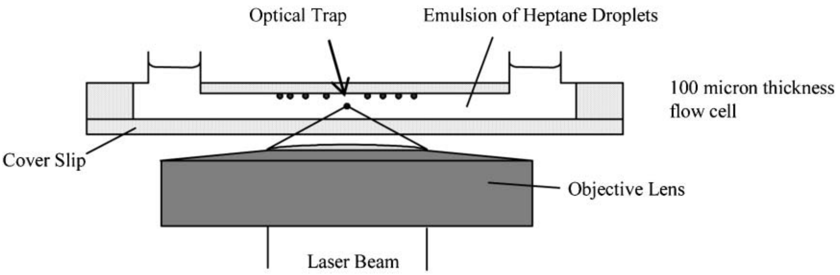

**Fig. 1** Schematic diagram of the flow cell illustrating the position of optical trapping and droplet deformation within the sample.

show the shape of the droplet as viewed in plan from beneath the sample using brightfield illumination. A side-on view of the droplet is not possible in the current optical configuration. To prevent the CCD from saturating with 1064 nm laser light, an optical filter was placed before the camera.

The maximum force exerted by a single optical trap was determined using an escape force technique, where a viscous drag force[10] was applied to a heptane droplet in the absence of salt, thus the oil–water interface was not in the ultralow region. As the solution flowed past the trapped droplet the laser power was reduced until the droplet just escaped. A linear relationship between laser power and the applied drag force was recorded, yielding an escape force of 0.69 pN $\text{mW}^{-1}$ for a single optical trap.

Fig. 2 shows an image of an oil droplet in a single optical trap (Fig. 2a) and a series of droplets deformed symmetrically by 2, 3 and 4 traps (Fig. 2b, c and d). With a single optical trap, the interfacial tension at the oil–water surface is sufficiently low such that the droplet deforms into an ellipse along the axial direction of the laser beam. This deformation is inferred from the observation that increasing laser power decreases the radius of the droplet and *vice versa*. The measured droplet radius as a function of laser power was extrapolated to zero power to obtain the spherical volume of the droplet. Across the range of laser powers used in this experiment the radius decreases by approximately 10%. Although we do not know the 3-dimensional shape of the structures formed with multiple optical traps, the average depth in the axial direction can be calculated on the reasonable assumption that the volume of the droplet remains constant throughout the deformation process. For example, the volume of the spherical droplet in a single trap with corrected radius, *r*, can be compared to that of an ellipsoid in the two trap deformation. The semi-major, *a*, and semi-minor, *b*, dimensions are estimated to the nearest 0.1 μm from the microscope images (1 pixel is 0.095 μm). From this assumption the semi-minor depth value, *c*, is equivalent to *b* within the experimental error of measurement. Thus, the droplet is deformed to a prolate spheroid with a small increase in surface area. The triangle and square droplet shapes appear to have a thicker central region that thins in the "corners". The contrast in the images does not suggest that there is a change from positive to negative curvature. In qualitative terms, the optical sculptures can be understood as photonic forces acting on the oil towards the focal points of the laser beam, balanced by a weak interfacial tension. The droplet assumes a shape of minimum surface area between the positions of the laser traps.

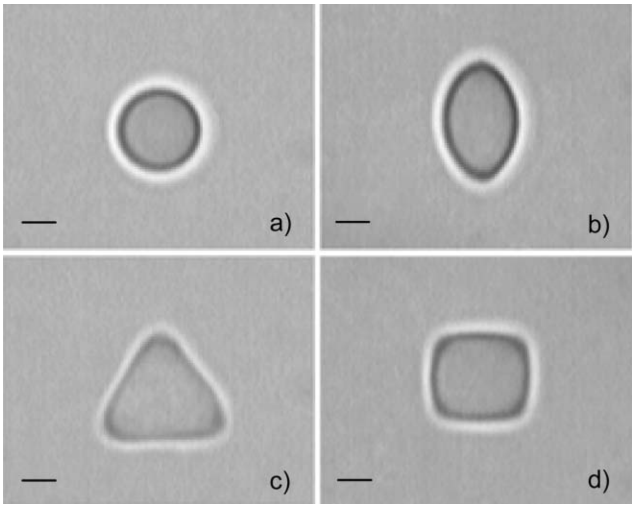

**Fig. 2** Shapes formed by manipulation of multiple optical traps inside an emulsion droplet: a) droplet in a single trap, b) ellipsoid formed by stretching the droplet with two traps, c) triangular shape formed using three traps, d) square shaped droplet formed with four traps. The images are in plan view and the scale bar represents 2 μm. Laser power is 24 mW divided between respective number of trapping positions.

A series of experiments were then performed in which a droplet was deformed into an ellipsoid, with two optical traps at a set laser power, until the surface area could not be extended further. If the trap positions were separated beyond this point, the interfacial elasticity overcame the optical trapping force and the stretched droplet dislodged from one of the traps. The droplet remained in the other trap and the traps could be reset to repeat the deformation. In this way, the maximum deformation could be measured as a function of the laser power (Fig. 3). Extending our geometry calculations enables a first order approximation of the interfacial tension of the droplets. The Laplace pressure arising from the curvature of the prolate spheroid is balanced by the applied force, *F*, over the droplet cross-section (eqn 1).

$$\frac{F}{\pi a^2} = \gamma\left(\frac{2}{R_1} - \frac{1}{a} - \frac{1}{b}\right) \qquad (1)$$

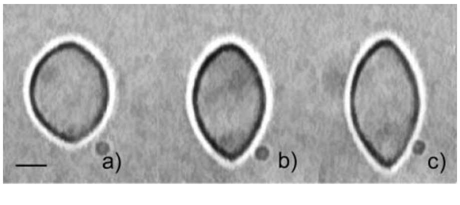

**Fig. 3** Limiting droplet extensions with two optical traps as a function of laser power: a) 11 mW, b) 19 mW, c) 22 mW. Scale bar represents 2 μm.

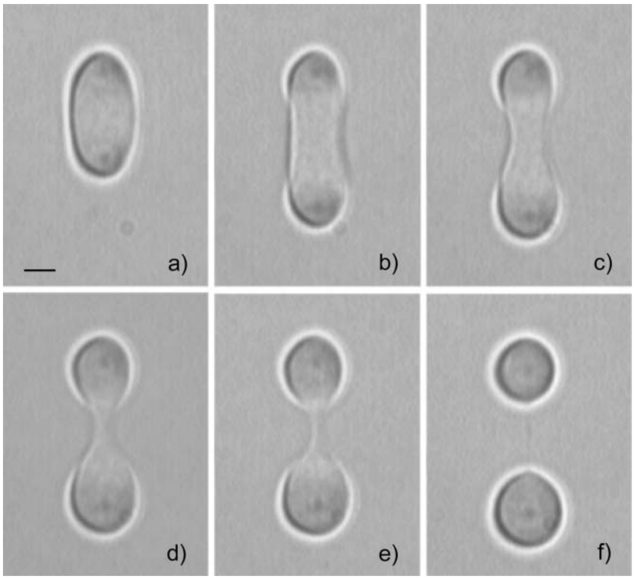

**Fig. 4** With very low tensions the droplets form dumbbell shapes on extension and can be separated and rejoined. Time elapsed between each image is 1 second. Careful inspection of image f) reveals that a thin thread connects the separated droplets. Laser power is 27 mW.

where: $R_1$ is the radius of curvature at the end of the droplet. From the escape force calibration, the optical force exerted at each node at the maximum extension was 8 pN; this value is only an estimate, since the shape of the interface through which the laser passes is no longer spherical. The values of $R_1$, $a$ and $b$ are approximately 1 μm, 2 μm and 5 μm, from which we estimate a value of the interfacial tension of $\sim 1 \times 10^{-6}$ N m$^{-1}$, which is in good agreement with the results obtained by Aveyard[7] and Mitani[6] using spinning drop interfacial tensiometry. The value of the interfacial tension depends sensitively on temperature. At present, we have fixed both temperature and salt concentration during the deformation experiments. However, varying these parameters will allow a better comparison of the interfacial properties determined from tensiometry and optical deformation techniques.

Under higher powers (27 mW), some droplets continued to stretch into a dumbbell shape (Fig. 4) which then divided into two smaller droplets as the trap separation was increased. These droplets appeared to remain connected by a thin oil thread barely discernable in the microscopic images and reminiscent of the ligament that connects a droplet to a nozzle in an inkjet printer. Closer inspection of the two droplets showed that they formed a point where the threads were attached, indicating the presence of a small tethering force. The two parts could readily be joined again, with no barrier to coalescence, to reform the initial droplet.

We have shown that it is possible to deform spherical emulsion droplets into a variety of shapes such as triangles and squares using multiple optical traps, provided that the interfacial tension, which is of the order of $10^{-6}$ N m$^{-1}$, is comparable to the force constant of the trap. Such low interfacial tensions can be achieved by working with oil–surfactant systems, such as heptane, Aerosol OT and brine, that are known to form microemulsions.

## Notes and references

1 *Methods in Cell Biology*, ed. M. P. Sheetz, Academic Press, San Diego, CA, USA, 1998, vol. 55.

2 P. J. H. Bronkhorst, G. J. Streekstra, J. Grimbergen, E. J. Nijhof, J. J. Sixma and G. J. Brakenhoff, *Biophys. J.*, 1995, **69**, 1666; J. Guck, R. Ananthakrishnan, T. J. Moon, C. C. Cunningham and J. Käs, *Phys. Rev. Lett.*, 2000, **84**, 5451.

3 A. Ashkin and J. M. Dziedzic, *Phys. Rev. Lett.*, 1973, **30**, 139.

4 J.-Z. Zhang and R. K. Chang, *Opt. Lett.*, 1988, **13**, 916.

5 A. Casner and J.-P. Delville, *Phys. Rev. Lett.*, 2001, **87**, 054503.

6 S. Mitani and K. Sakai, *Phys. Rev. E: Stat. Phys., Plasmas, Fluids, Relat. Interdiscip. Top.*, 2002, **66**, 031604.

7 R. Aveyard, B. P. Binks, S. Clark and J. Mead, *J. Chem. Soc., Faraday Trans.*, 1986, **82**, 125.

8 C. D. Mellor, M. A. Sharp, C. D. Bain and A. D. Ward, *J. Appl. Phys.*, 2005, **97**, 103114.

9 E. J. G. Peterman, F. Gittes and C. F. Schmidt, *Biophys. J.*, 2003, **84**, 1308.

10 R. Aveyard, B. P. Binks, J. H. Clint, P. D. I. Fletcher, T. S. Horozov, B. Neumann, V. N. Paunov, J. Annesley, S. W. Botchway, D. Nees, A. W. Parker, A. D. Ward and A. N. Burgess, *Phys. Rev. Lett.*, 2002, **88**, 246102.