

Improved optically driven microrotors

Theodor Asavei, Vincent L. Y. Loke, Timo A. Nieminen, Norman R. Heckenberg,
Halina Rubinsztein-Dunlop
Centre for Biophotonics and Laser Science, School of Physical Sciences, The University of
Queensland, Brisbane QLD 4072, Australia

ABSTRACT

Two-photon polymerization of optically curing resins is a powerful method to fabricate micron sized objects which can be used as tools to measure properties at small scales. These microdevices can be driven by means of externally applied focused laser beams (optical tweezers) through angular momentum exchange, giving rise to a net torque. The advantage of the optical drive is that no contact is required, therefore making the microdevices suited to non-invasive biological applications.

The fabrication method is versatile and allows building objects of any 3D shape.

We discuss the design and modeling of various optically driven rotors. In particular, we consider fabrication of microspheres with an internal shape birefringence in order to obtain rotation in an optical trap. The reason for fabricating this type of object is that they are well-suited for studies of mechanical properties of single biomolecules such as the torsional stiffness of DNA or torque generated by molecular motors.

Keywords: Two-photon photopolymerization, optical trapping, optical angular momentum

1. INTRODUCTION

Micromachines are micron-sized devices which can be used as tools for investigating a large number of phenomena at small scale. Therefore the fabrication of such objects is extremely important.

Richard Feynman anticipated the idea of micromachines in his 1959 talk, "There's plenty of room at the bottom", published in 1960.^[1] He suggested as an example of a possible micromachine, the micromechanical "surgeon" which could be swallowed and operate inside a faulty blood vessel for example. He also pointed out how physics of machines at the microscale would be different than for macroscale machines. At the end of his talk Feynman offered two prizes, one for building an operating electric motor only the size of a 1/64 inch cube and the other for writing a page with letters 1/25,000 smaller than in a normal text. It didn't take long until the first micromachine (the 1/64 inch cube motor) was built in November 1960 by James McLellan, an electrical engineer.

The second prize was won in 1985 by Tom Newman, a Stanford graduate student, who wrote the first page of "A Tale of Two Cities" in polymethyl methacrylate (PMMA) resist on a silicon nitride membrane by means of electron beam lithography.^[2,3]

At the end of the 1980s and the beginning of the 1990s, microelectromechanical systems (MEMS) came into play. MEMS, commonly known as micromachines, are electromechanical machines that range in size from a micrometer to a millimetre. They are built using techniques based on integrated circuit (IC) fabrication methods.

The first reported MEMS were silicon electrical micromotors with a diameter of 100 μm .^[4,5]

Nowadays MEMS are used as sensors and actuators in various applications. Some of the common applications include accelerometers that trigger airbags in cars, inkjet printers, blood pressure sensors or optical switching for data communications. Although not yet very common, MEMS are finding their ways into medical application.^[6-8] However, MEMS are not the only available micromachines. Another type of micromachine is the optically fabricated type. They emerged as a consequence of the rapid prototyping techniques which became available in the 1980s, namely the stereolithography technique.^[9]

There are several methods of producing micromachines. The oldest one is bulk micromachining [10], and it is used to fabricate silicon based MEMS. This method creates micro-mechanical structures by etching into a silicon wafer, using the whole thickness of the wafer.

As opposed to bulk micromachining, surface micromachining^[10] was created in the late 1980s in order to obtain planar MEMS which could be compatible with on-chip integrated circuits. This technique creates MEMS elements from thin

films deposited on the surface of a silicon wafer. Polycrystalline silicon, silicon nitride or silicon dioxide films can be deposited layer-by-layer on the silicon wafer, photo-patterned with photolithography and selectively removed through etching to obtain the desired machine elements. However, a drawback of surface micromachining is the fact that it is limited to the production of two-dimensional structures. In other words one cannot create three-dimensional structures with high aspect ratio, or curved surfaces with surface micromachining. Apart from the above mentioned methods, micromachines can be fabricated optically by means of microstereolithography. This method has the advantage that it can create real 3D microstructures. It is based on the fact that certain resins can be hardened by exposure to UV light. Basically the 3D microstructure is fabricated by stacked 2D hardened elements, which are initially obtained from slicing the 3D shape of the structure in a computer-aided design (CAD) program.

The fact that some resins harden under UV exposure is based on photopolymerization of monomers in the liquid resin. The photopolymerization process is a particular type of radical chain polymerization. Radical chain polymerization is a polymerization mechanism in which an initiator molecule produces a reactive centre in the form of a free radical. Polymerization occurs as a chain reaction by successive addition of monomers to the reactive centre. The chain reaction consists of three steps: initiation, propagation and termination.^[11] The initiation step consists of two reactions. The first reaction is the homolytic dissociation of the initiator molecule, I, to obtain a pair of free radicals, R[•]. In the case of photopolymerization, the dissociation is produced by absorption of light photons. The second reaction of initiation is the addition of the radical to the first monomer molecule, M, producing the chain-initiating radical M₁[•].

The propagation step consists of the successive additions of monomer molecules to M₁[•].

Termination occurs when the radical centres are annihilated by combination (coupling) reaction between radicals. M_{n+m} also referred to as the dead polymer (the hardened resin) is the final product of the photopolymerization process.

The first optically fabricated microstructures reported in 1993 were a micro valve, a micro coil spring with a 250 μm height and 50 μm coil diameter and a micro pipe with 30 μm inner diameter.^[12] They were fabricated with a Xenon lamp as UV source focused in a liquid resin. The resin hardened only in the focal spot of the UV beam and by scanning the sample over the focus 3D objects could be obtained.

The resolution of the optically fabricated microstructures is determined by the size of the smallest solidified volume, called "voxel" (volumetric pixel). For the above mentioned objects the resolution was 5x5x3 μm³. The resolution can be substantially increased if instead of using one-photon absorption of UV light one uses two-photon absorption of IR light. The increase in resolution is due to the fact that two-photon absorption probability is proportional to the square of light intensity and hence the resin polymerizes in a far smaller volume than the one in one-photon absorption.

The two-photon polymerization technique was pioneered by J. Strickler and W. Webb in 1991, following the application of two-photon excitation in two-photon laser scanning fluorescence microscopy.^[13] Two-photon excitation was used to record high-density digital information in a multilayered 3D format. The information was written in submicrometre size voxels in a photopolymer^[14] with femtosecond IR laser pulses.

The first 3D microfabricated structures with two-photon polymerization were reported in 1997.^[15] They were spiral structures with a diameter of 6 μm and a wire width of 1.3 μm. Since then, various micromachines have been produced (micropumps, microgears, microneedles) with high resolution.^[16-18] A lot of research is being done in order to improve the spatial resolution of the photopolymerization process so that objects with fabrication accuracy of 150 nm can now be produced.^[19]

The most common drive mechanism encountered in surface-micromachined silicon based elements is the electrical drive. This is achieved by means of electrostatic forces which convert electrical energy to mechanical energy. The basic elements of the electrical driven micromachine are the stator and the rotor. By applying a three-phase voltage difference between the stator and the rotor one can achieve continuous motion of the rotor. However, micromachines can also be driven by thermal energy through thermal expansion,^[20] by magnetic fields,^[21] by chemical energy^[22] or by a tightly focussed laser beam (optical drive).^[23-25]

Optical drive is based on the fact that microscopic objects (25 nm to 10 μm) can be stably trapped in a tightly focused laser beam due to gradient forces. The single-beam gradient optical trap, also called optical tweezers, was first achieved by Ashkin et al. in 1985.^[26] Forces on the order of pN with mW of laser power can be exerted on the trapped particles.

The advantage of optical drive is that no contact is required so that it is particularly well suited to biological applications. This type of drive is particularly suited to optically fabricated micromachines because they are made out of transparent polymers. However microstructures built by surface micromachining can be optically driven too.

The earliest reported optically driven micro-objects were fabricated by means of reactive ion-beam etching of a 10 micron thick silicon dioxide (SiO₂) layer^[23] resulting in 10 to 25 μm diameter microrotors which could be rotated in the optical trap.

If the micromachines are to be used in biological applications they need to be made out of biocompatible materials and have a size which can be easily tailored to the studied systems. Here, our micromachines are fabricated by two-photon polymerization of optically curable resins, which fulfil the above requirements.

2. RESULTS AND DISCUSSION

There has been an ongoing effort during the past decade to manipulate and probe individual biological molecules (DNA, molecular motors).^[27-29] In this type of study forces and torques exerted on single-molecules are measured in order to understand processes at cellular level in which these molecules are involved. In order to perform these experiments one end of the molecule has to be attached to a fixed surface and the other to a micrometer sized object that could be easily optically trapped. The most common particles used in optical trapping are isotropic spheres which are only suitable for force measurements and translational motion. However, rotation of biomolecules plays a big role in the cellular mechanisms (DNA supercoiling, DNA transcription, DNA repair). In order to measure torques in an optical trap one needs either optical or shape birefringent particles.

Rotation measurements have been performed using optically birefringent calcium carbonate crystal (vaterite) microspheres. However these spheres are not very stable in the biological buffer environment. Recently a new type of micrometer sized objects were reported, namely quartz microcylinders for DNA torque detection fabricated by standard photolithography.^[30]

We propose here a polymer microsphere fabricated by two-photon polymerization for torque measurements. Due to its symmetry a homogenous isotropic sphere does not rotate in an optical trap. However one could break the symmetry by designing a sphere with a cavity inside which can induce shape birefringence. One can then optimize the angular momentum transfer between the laser beam and the object, and hence the torque, by changing the shape of the cavity which can be easily done with the two-photon polymerization technique.

We have designed and fabricated microspheres with a cylindrical rod-shaped void inside. As discussed in the experimental section, the 3D computer designed object (figure 1A) is sliced into bitmaps (figure 1B) which are then input into the scanning program. Due to the versatility of the technique we were able to vary the diameter of the spheres as well as the diameter of the void. An optical microscope image of 4 μm diameter spheres with various void sizes in unpolymerized resin is shown in figure 1C. Due to the high travel range of the scanning stage one could produce a large number of microstructures in one go which is a big advantage in terms of fabrication efficiency. The fabrication time for each structure is about 25 minutes. The structures were also imaged with the scanning electron microscope (SEM), which is a powerful tool due to its high resolution in the nanometer range. A typical SEM image of the fabricated structure is shown in figure 1D. The layer-by-layer formation of the microstructure can be clearly seen from the SEM image.



Figure 1 CAD model of the microfabricated sphere (A), 2D bitmaps used to fabricate the micro-object (B), bright field microscope image of 4 μm diameter spheres (C) and SEM image of a 10 μm diameter sphere with 1 μm hollow rod (D).

We have also studied the spatial resolution of the polymerization process which is given by the size of a single voxel. Theoretically the voxel is a prolate spheroid and this follows from the calculation of the diffraction limited spot for the

two-photon process which has this shape. Theoretically it is shown^[31] that for a 1.3 N.A. objective lens the ratio between the long axis of the spheroid (which determines the axial resolution) and the short axis (the lateral resolution) is 3:1. Even though the shape of the single voxel sets a low axial resolution one can often increase the axial resolution by overlapping layers in the Z direction. Experimentally it is found that the resolution is dependent on the exposure time as well as the power of the laser beam.

The resolution of the two-photon polymerization process was investigated by producing individual voxels at different exposure times at a given power. In figure 2 is shown a SEM image of a single voxel produced at 20 mW and 60 ms exposure time. It has a diameter of 270 nm and a length of 1.4 μm.

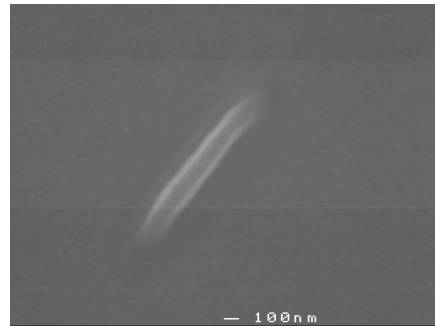


Figure 2 SEM image of a single voxel.

At the same power, a series of voxels were produced with different exposure times ranging from 40 ms to 500 ms, the smallest voxel having a diameter of 210 nm and a length of 970 nm. However, when fabricating an object some features below those limits can be generated by overlapping subsequent layers..

Torque measurements, as described in the experimental section, were performed in circularly polarized light on 4 μm diameter microspheres attached to the cover glass. These microstructures were fabricated with the cylindrical void perpendicular to the direction of propagation of the laser beam. The measured torque efficiency of the microspheres as a function of the void diameter is shown in figure 3. In these measurements we find the torque efficiency in the order of 0.02 ħ per photon. One drawback of this type of experiment is the fact that the rod was fabricated horizontally and hence the actual size could be smaller than the designed one. Therefore higher torques can be expected when performing an experiment with a detached microsphere with a vertical void.

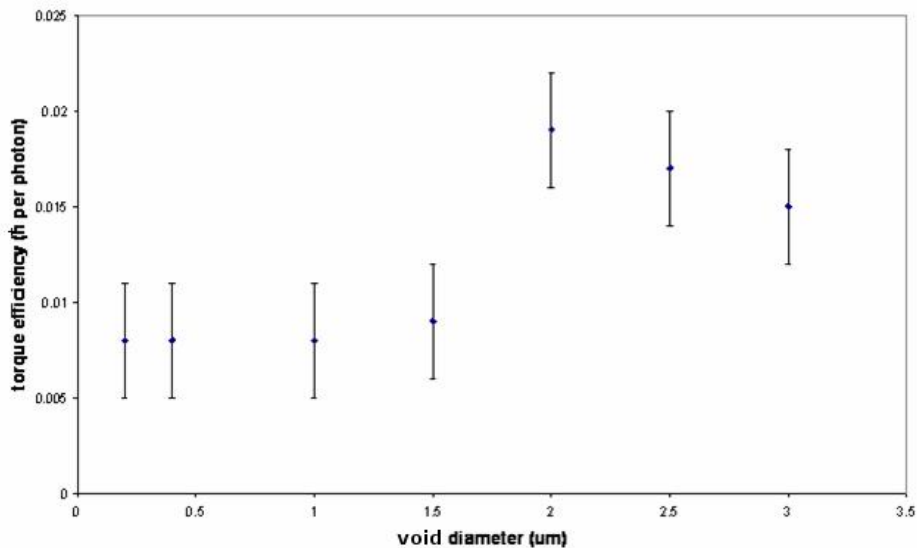


Figure 3 Torque efficiency of the microspheres as a function of void diameter.

After fabrication, the microspheres could be detached from the cover slip and used for optical trapping experiments. The sphere oriented itself with the rod perpendicular to the direction of propagation of the trapping beam and started to rotate in circularly polarized light. In figure 4 are shown three consecutive snapshots from a rotating sphere experiment. The rotation frequency is 10 Hz with 250 mW incoming power of the trapping beam.

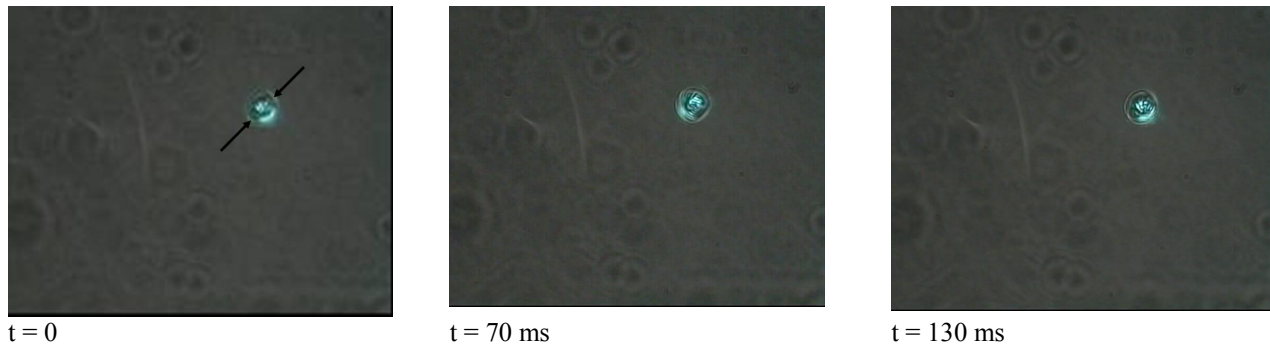


Figure 4. Bright field microscope image of a rotating microsphere. The arrows point towards the direction of the void.

3. EXPERIMENTAL DETAILS

We use the UV curing resin NOA81 (Norland Products Inc., Cranbury, NJ, USA) to produce the microstructures. This resin is based on a mixture of photoinitiator molecules and thiol-ene monomers. The resin is photopolymerized when exposed to light in the UV range with $\lambda < 400$ nm and require an energy flux of 2 to 4.5 J/cm² for a full cure (NOA81 technical data sheet).

The two-photon polymerization is performed in an “in house” built inverted microscope. A schematic of the setup is shown in figure 5.

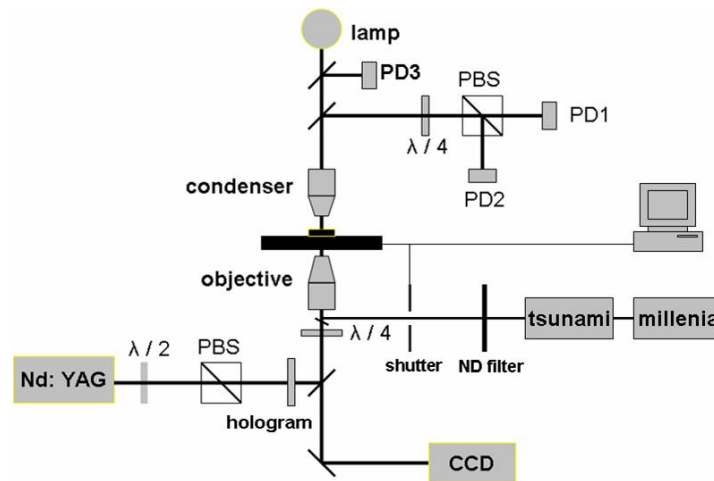


Figure 5 Schematic of the experimental setup.

For two-photon polymerization we use the infrared light ($\lambda=780$ nm) produced by a femtosecond pulse Ti:Sapphire laser (Tsunami, Spectra Physics Inc.) pumped by a 532 nm solid state laser (Millenia, Spectra Physics Inc.). The pulse length is 80 fs with 80 MHz repetition rate.

The laser beam is attenuated by a rotating variable optical density filter wheel to the power needed for polymerization (20 mW average power measured before entering the microscope) and then passes through a computer controlled shutter and is reflected into the objective lens by a dichroic mirror. The objective lens is an Olympus 100X oil immersion lens with high numerical aperture (N.A. =1.3) to achieve high spatial resolution for polymerization.

The sample is mounted on a computer controlled piezo actuated stage (model P-611.3S, PI, Physik Instrumente) and is imaged onto a CCD with the same objective lens. The travel range of the piezo stage is 100 μm in all X, Y and Z directions.

The resin sample is sandwiched between two glass cover slips which are separated by an adhesive spacer (Parafilm, Structure Probe Inc., West Chester, PA, USA) with a thickness of 127 μm . 3D structures are fabricated by raster scanning the resin sample over the laser beam using the piezo stage. A schematic of the fabrication method is shown in figure 6.

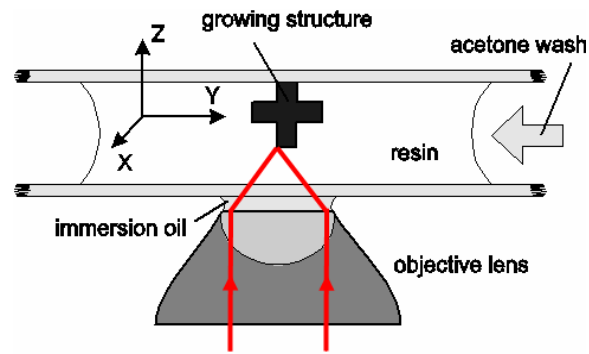


Figure 6 Schematic of the microfabrication method.

The 3D object is sliced into 2D layers (bitmap files) corresponding to the areas that need to be scanned. The program controlling the scanning stage reads the bitmap files and the resin is exposed (the shutter is opened) when the pixel in the bitmap is black and the shutter closes when the pixel is white. 3D structures are obtained by moving the sample in the Z direction after each XY scan.

The bitmap resolution is set to 100x100 pixels which corresponds to 10x10 μm^2 travel in X and Y directions so that each individual pixel is 100x100 nm^2 in size. The offset step in the Z direction is 200 nm.

The structures are grown upside down on the upper cover slip. This top down scanning method has the advantage that the laser beam does not pass through already exposed resin.

After the polymerization, the unexposed resin is washed off with acetone, leaving the 3D structure attached to the cover slip.

Optical trapping is performed in an inverted microscope built "in house" (figure 5). The trapping laser is a 5 W, 1064 nm Nd:YAG fiber laser (IPG Photonics, Oxford, MA, USA). It is focused on the sample by a 100X Olympus oil immersion objective lens with high numerical aperture (N.A. = 1.3). The output power is controlled by a half wave plate ($\lambda/2$) and a polarizing beam splitter cube (PBS). Circular polarization of the trapping beam is obtained by using a quarter wave plate ($\lambda/4$). The sample is imaged by using the same objective lens.

In order to detach the produced microstructures from the cover slip we use a micropipette mounted on a XYZ stage. The tip of the micropipette is 2 μm in diameter. The microstructures can be easily detached by touching them with the pipette tip. The freed structure is then trapped in optical tweezers.

The microspheres with the void inside, due to their induced form birefringence, will change the polarization state of an incident circularly polarized beam and hence a torque will be exerted on them. In other words the torque is due to the spin angular momentum transfer. A schematic of the process is shown in figure 7.

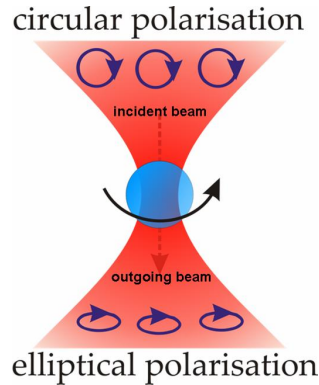


Figure 7 Cartoon showing the change in polarization due to form birefringence.

It can be shown^[32] that any beam can be represented as a sum of two circularly polarized components with opposite handedness with a coefficient of circular polarization σ given by $\sigma = (P_L - P_R) / P$, where P_L and P_R are the powers of the left and right circularly polarized components respectively and P is the total power of the beam. Therefore the torque τ can be written as $\tau = \Delta\sigma P / \omega$,^[32] with $\Delta\sigma$ being the change in the coefficient of circular polarization due to the spin angular momentum transfer, P is the incident beam power and ω the beam angular frequency. Hence by measuring $\Delta\sigma$ and knowing P and ω , the torque can be found. The change in the coefficient of circular polarization is measured by two photo detectors (PD) which are placed after a polarizing beam splitter cube (figure 5). The outgoing beam is collimated by the condenser and then split into two orthogonal linearly polarized components by the quarter wave plate ($\lambda / 4$) and the cube. The two linearly polarized components correspond to the left and right circularly polarized components of the outgoing beam. The two detectors measure the power of each beam and hence the coefficient $\Delta\sigma$ can be found.

The microstructures are characterized by electron microscopy. The cover slips with the produced structures are mounted on an adhesive carbon pad and then coated with a 10 nm platinum layer. We use a field emission scanning electron microscope (JEOL JSM-6300F) operated at 15 kV.

4. MODELING

In order to calculate the expected force and torque on a particular microfabricated object, we employ a modeling method known as discrete dipole approximation (DDA) [33], whereby the target object is represented by a dipole lattice (figure 8). The dipole fields are induced by the incident beam and the interaction between the other dipoles. The dipole moments are obtained by solving a number of linear equations equivalent to the number of dipoles in three dimensions. Subsequently, the forces and torques can be calculated via a variety of methods, including finding the force on each individual dipole [34,35], or even by calculating a T-matrix for the scatterer [36].

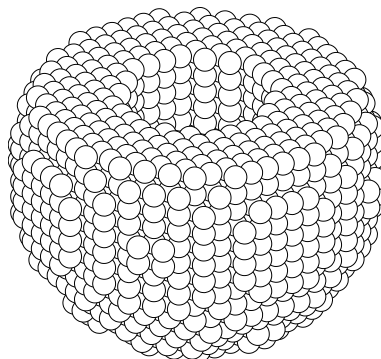


Figure 8 Dipole lattice model of a microfabricated object.

5. CONCLUSION

Two-photon photopolymerization of optically curable resins is a powerful method of fabricating 3D micrometer size structures of arbitrary shape. The high lateral resolution of this process, below the diffraction limited resolution of light, allows fabrication of micro-objects with fine features. Even though the axial resolution of the process is inherently lower than the lateral resolution, it can be reduced by setting a small offset between subsequent layers.

We have shown the applicability of the method by fabricating micro-structures with induced form birefringence which can rotate in an optical trap with circular polarization. These objects can be used in studies of mechanical properties of biological molecules such as torsional and elastical properties of DNA or molecular motors.

REFERENCES

- [1] R. P. Feynman, "There's plenty of room at the bottom", *California Institute of Technology Journal of Engineering and Science* **1960**, 4, 23-36.
- [2] "Tiny tale gets grand", *California Institute of Technology Journal of Engineering and Science* **1986**, 49, 24-28.
- [3] T. H. Newman, K. E. Williams, R. F. W. Pease, "High resolution patterning system with a single bore objective lens", *Journal of Vacuum Science and technology B* **1987**, 5, 88-91.
- [4] L. S. Fan, Y. C. Tai, R. S. Muller, "IC-processed electrostatic micromotors", *Sensors and Actuators* **1989**, 20, 41-47.
- [5] M. Mehregany, S. F. Bart, L. S. Tavrow, J. H. Lang, S. D. Senturia, M. F. Schlecht, "A study of three microfabricated variable-capacitance motors", *Sensors and Actuators A* **1990**, 21, 173-179.
- [6] E. Benzel, L. Ferrara, S. Roy, A. Fleishman, "Micromachines in spine surgery", *Spine* **2004**, 29, 601-606.
- [7] K. Ishiyama, M. Sendoh, K. I. Arai, "Magnetic micromachines for medical applications" *Journal of Magnetism and Magnetic Materials* **2002**, 242, 41-46.
- [8] P. Goh, S. M. Krishnan, "Micromachines in endoscopy" *Bailliere's Clinical Gastroenterology* **1999**, 13, 49-58.
- [9] D. Deitz, "Stereolithography automates prototyping", *Mechanical Engineering* **1990**, 112, 34-39.
- [10] D. Lopez, F. Pardo, C. Bolle, R.S. Decca, D. Bishop, , "MEMS Technology for the Advancement of Science" *Journal of Low Temperature Physics* **2004**, 135, 51-62.
- [11] G. Odian, *Principles of polymerization*, John Wiley & Sons, **2004**.
- [12] K. Ikuta, K. Hirowatari, "Real three dimensional micro fabrication using stereo lithography and metal moulding", *Proceedings of the IEEE Workshop on MEMS* **1993**, 42-47.
- [13] W. Denk, J. H. Strickler, W. W. Webb, , "Two-photon laser scanning fluorescence microscopy", *Science* **1990**, 248, 73-76.
- [14] J. H. Strickler, W. W. Webb, "Three-dimensional optical data storage in refractive media by two-photon point excitation", *Optics Letters* **1991**, 16, 1780-1782.
- [15] S. Maruo, O. Nakamura, S. Kawata, "Three-dimensional microfabrication with two-photon-absorbed photopolymerization" *Optics Letters* **1997**, 22, 132-134.
- [16] S. Maruo, H. Inoue, "Optically driven micropump produced by three-dimensional two-photon microfabrication", *Applied Physics Letters* **2006**, 89, 144101.
- [17] S. Maruo, K. Ikuta, H. Korogi, "Submicron manipulation tools driven by light in a liquid", *Applied Physics Letters* **2003**, 82, 133-135.
- [18] P. Galajda, P. Ormos, "Complex micromachines produced and driven by light" *Applied Physics Letters* **2001**, 78, 249-251.
- [19] S. Kawata, H. B. Sun, T. Tanaka, K. Takada, "Finer features for functional microdevices", *Nature* **2001**, 412, 697-698.
- [20] A. D. Oliver, S. R. Vigil, Y. B. Gianchandani, "Photothermal surface-micromachined actuators", *IEEE Transactions on Electron Devices* **2003**, 50, 1156-1157.
- [21] K. Nishimura, H. Uchida, M. Inoue, M. Sendoh, K. Ishiyama, K. I. Arai, "Magnetic micromachines prepared by ferrite plating technique", *Journal of Applied Physics* **2003**, 93, 6712-6714.
- [22] Y. Hiratsuka, M. Miyata, T. Tada, T. Q. P. Uyeda, "A microrotary motor powered by bacteria", *PNAS* **2006**, 103, 13618-13623.
- [23] E. Higurashi, H. Ukita, H. Tanaka, O. Ohguchi, "Optically induced rotation of anisotropic micro-objects fabricated by surface micromachining", *Applied Physics Letters* **1994**, 64, 2209-2210.
- [24] M. E. J. Friese, H. Rubinsztein-Dunlop, J. Gold, P. Hagberg, D. Hanstorp, "Optically driven micromachine elements", *Applied Physics Letters* **2001**, 78, 547-549.

- [25] L. Kelemen, S. Valkai, P. Ormos, *Applied Optics* **2006**, 45, 2777-2780.
- [26] A. Ashkin, J. M. Dziedzic, J. E. Bjorkholm, S. Chu, "Observation of a single-beam gradient force optical trap for dielectric particles", *Optics Letters* **1986**, 11, 288-290.
- [27] S. B. Smith, L. Finzi, C. Bustamante, "Direct mechanical measurements of the elasticity of single DNA molecules by using magnetic beads", *Science* **1992**, 258, 1122-1126.
- [28] W. S. Ryu, R. M. Berry, H. C. Berg, "Torque generating units of the flagellar motor of E-coli have a high duty ratio", *Nature* **2000**, 403, 444-447.
- [29] L. Oroszi, P. Galajda, H. Kirei, S. Bottka, P. Ormos, "Direct measurement of torque in an optical trap and its application to double-strand DNA", *Physical Review Letters* **2006**, 97, 058301.
- [30] C. Deufel, S. Forth, C. R. Simmons, S. Dejgosh, M. D. Wang, *Nature Methods* **2007**, 4, 223-225.
- [31] G. Knöner, J. Higuete, S. Parkin, T. A. Nieminen, N. R. Heckenberg, H. Rubinsztein-Dunlop, "Two-photon polymerization process for optically driven micromachines", *Proc. SPIE* **2006**, 6038, 208-216.
- [32] T. A. Nieminen, N. R. Heckenberg, H. Rubinsztein-Dunlop, "Optical measurement of microscopic torques", *Journal of Modern Optics* **2001**, 48, 405-413.
- [33] B. T. Draine, P. J. Flatau, "Discrete-dipole approximation for scattering calculations", *J. Opt. Soc. Am. A* **1994**, 11, 1491-1499.
- [34] Y. Harada, T. Asakura, "Radiation forces on a dielectric sphere in the Rayleigh scattering regime", *Optics Communications* **1996**, 124, 529-541.
- [35] A. G. Hoekstra and M. Frijlink and L. B. F. M. Waters, P. M. A. Sloot, "Radiation forces in the discrete-dipole approximation", *J. Opt. Soc. Am. A* **2001**, 18, 1944-1953.
- [36] D. W. Mackowski and M. I. Mishchenko, "Calculation of the T -matrix and the scattering matrix for ensembles of spheres", *J. Opt. Soc. Am. A* **1996**, 13, 2266-2278.