20181009 Masuhara-Sugiyama Joint Lab Meeting

2018 Nobel Prize Physics Awardee Dr. Arthur ASHKIN and Our Research on Laser Trapping Dynamics and Chemistry

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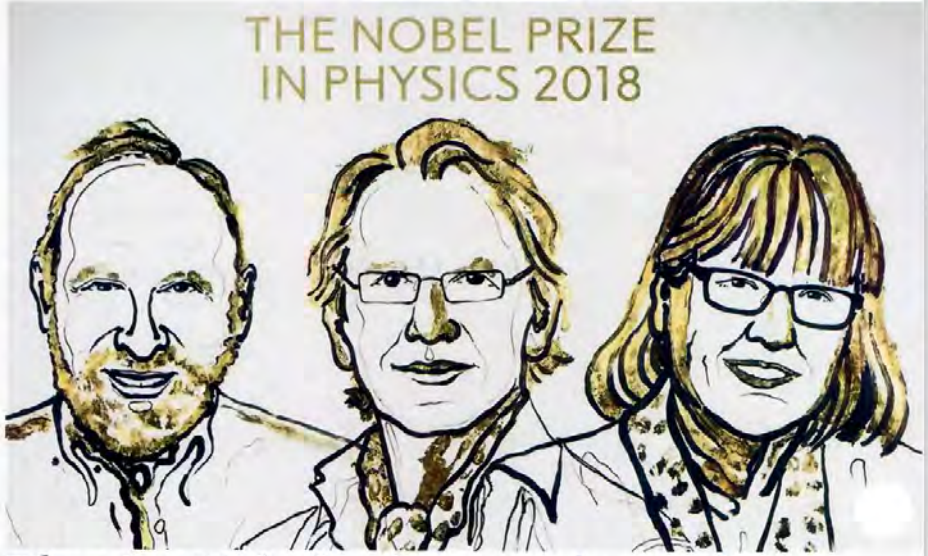
The Guardian



Physics Nobel prize won by Arthur Ashkin, Gérard Mourou and Donna Strickland

American, Frenchman and Canadian share 9m Swedish kronor (£770,000) prize for work in laser physics

Ian Sample *and* **Nicola Davis**

Tue 2 Oct 2018 11.15 BST

Three scientists have been awarded the 2018 Nobel prize in physics for creating groundbreaking tools from beams of light.

proportion of women who have won the physics prize. "That's why we are taking measures to encourage more nominations because we don't want to miss anyone."

Ashkin, who at 96 is the oldest winner of any Nobel prize, told the committee that he may not be able to give any interviews because he was "very busy" on his latest scientific paper. He had previously complained of being overlooked for the Nobel prize in 1997 when another Bell Labs researcher, the US physicist Steven Chu, shared the award for cooling and trapping atoms with lasers.

Through his research in the 1970s and 80s, Ashkin showed how the radiation pressure of light could be harnessed to move physical objects without burning them, realising "an old dream of science fiction," according to the Swedish Academy. In 1987, Ashkin published a landmark paper showing how optical tweezers could capture living bacteria without harming them.

Around the same time, Mourou and Strickland demonstrated how to create ultrashort, intense bursts of laser light by stretching, amplifying and finally recompressing light waves. Mourou said he had a key insight into solving the problem while riding on a ski lift at Bristol Mountain resort in Rochester, New York. The pair's seminal paper in 1985 was Strickland's first as an academic.

Amanda Wright, associate professor at the faculty of engineering, University of Nottingham, said she was "delighted" with the prize announcement. She said a recently-funded collaboration between Nottingham, Heriot Watt and the University of Glasgow will use optical tweezers to explore how cells talk to their surroundings and vice versa, "and how these interactions affect disease progression."

Jim Al-Khalili, professor of theoretical physics at the University of Surrey, added: "The most thrilling thing for me is to see Donna Strickland share this year's prize. It is quite shocking to know that she is only the third woman to win a physics Nobel ever," said "It

Arthur Ashkin
From Wikipedia, the free encyclopedia

Arthur Ashkin (born September 2, 1922)[1][2][3] is an American scientist and Nobel laureate who worked at Bell Laboratories and Lucent Technologies. Ashkin has been considered by many as the father of the topical field of optical tweezers,[4][5][6] for which he was awarded the Nobel Prize in Physics 2018. He resides in Rumson, New Jersey.[7]

Ashkin started his work on manipulation of microparticles with laser light in the late 1960s which resulted in the invention of optical tweezers in 1986. He also pioneered the optical trapping process that eventually was used to manipulate atoms, molecules, and biological cells. The key phenomenon is the radiation pressure of light; this pressure can be dissected down into optical gradient and scattering forces.

At Bell Labs, Ashkin worked in the microwave field until about 1960 to 1961, and then switched to laser research. His research and published articles at that time pertained to nonlinear optics, optical fibers, parametric oscillators and parametric amplifiers. Also, at Bell Labs during the 1960s, he was the co-discoverer of the photorefractive effect in the piezoelectric crystal.[4][18]

Ashkin's work formed the basis for Steven Chu's work on cooling and trapping atoms, which earned Chu the 1997 Nobel Prize in physics.[4][5]
Besides optical tweezers, Ashkin is also known for his studies in photorefraction, second harmonic generation, and non-linear optics in fibers.[4][5]
Recent advances in physics and biology using optical micromanipulation include achievement of Bose–Einstein condensationin atomic vapors at submillikelvin temperatures, demonstration of atom lasers, and detailed measurements on individual motor molecules.[4][5]

On October 2, 2018, Ashkin was awarded the Nobel Prize in Physics for his work on optical trapping.[21] Ashkin "was honored for his invention of 'optical tweezers' that grab particles, atoms, viruses and other living cells with their laser beam fingers. With this he was able to use the radiation pressure of light to move physical objects, 'an old dream of science fiction', the Royal Swedish Academy of Sciences said."[22]

Arthur Ashkin

Bell Labs retired

Laser trapping solar power

自分のプロフィールを作成

	すべて	2013 年以来
引用	37136	9967
h 指標	62	35
i10 指標	115	58

タイトル	引用先	年
Observation of a single-beam gradient force optical trap for dielectric particles A Ashkin, JM Dziedzic, JE Bjorkholm, S Chu Optics letters 11 (5), 288-290	6274	1986
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Forces of a single-beam gradient laser trap on a dielectric sphere in the ray optics regime A Ashkin Biophysical journal 61 (2), 569-582	1804	1992

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Optical trapping and manipulation of neutral particles using lasers A Ashkin Proceedings of the National Academy of Sciences 94 (10), 4853-4860	1622 *	1997
Optical trapping and manipulation of neutral particles using lasers A Ashkin Proceedings of the National Academy of Sciences 94 (10), 4853-4860	~~1388~~	1997
OPTICALLY-INDUCED REFRACTIVE INDEX INHOMOGENEITIES IN $LiNbO_3$, AND $LiTaO_3$ A Ashkin, CD Boyd, JM Dziedzic, RG Smith, AA Ballman, JJ Levinstein, ... Landmark Papers On Photorefractive Nonlinear Optics, 29-31	1326	1995
Three-dimensional viscous confinement and cooling of atoms by resonance radiation pressure S Chu, L Hollberg, JE Bjorkholm, A Cable, A Ashkin Physical review letters 55 (1), 48	1301	1985
Experimental observation of optically trapped atoms S Chu, JE Bjorkholm, A Ashkin, A Cable Physical review letters 57 (3), 314	1176	1986
Optical levitation by radiation pressure A Ashkin, JM Dziedzic Applied Physics Letters 19 (8), 283-285	809	1971
Motion of atoms in a radiation trap JP Gordon, A Ashkin Physical Review A 21 (5), 1606	766	1980

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1994年
出版

コラム・マイクロ化学あれこれ

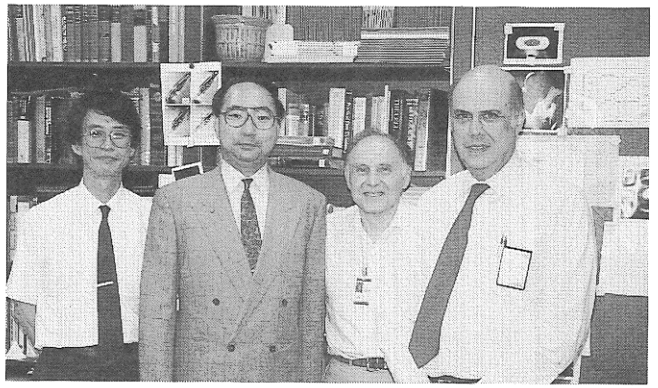
レーザーマニピュレーションの創始者

Ashkin 博士訪問記

1990年夏，著者ら(喜多村，三澤，笹木)は，英国で開催される IUPAC の光化学の学会に参加する機会を利用し，米国 AT&T Holmdel 研究所の Ashkin 博士を訪問する計画を立てた．本文でも触れたように，Ashkin 博士は世界で初めて微粒子のレーザーマニピュレーションに成功し，その後も関連する革新的な研究を進められた方である．博士の研究室をぜひ訪問したいと考えたのは，世界的な権威である博士とわれわれの研究についてディスカッションしておくことが，今後，レーザーマニピュレーションを用いた研究を進めるうえでわれわれの大きな自信につながると確信したからである．しかし，まったく面識のない，しかも物理ではなく化学の分野の研究者がいきなり訪問したいと手紙を書いても，会っていただけないのではないかとの危惧があった．そこで光化学の研究を通じて増原先生と親交のある，同じ AT&T Murray Hill 研究所の Chandross 博士を通じて訪問の打診を行ったところ，快くお引き受けいただくことができた．

著者らは英国の学会が終わった後，心を弾ませながら大西洋を渡り，ニュージャージー州のニューアーク飛行場に着いた．その翌日，飛行場近くのレンタカー屋から赤いムスタングを借りて，一路 AT&T Holmdel 研究所へ向かったのである．高速道路を降りて緑の多い閑静な住宅地のなかを抜けていくと，突然，鏡張りの大きな研究所のビルディングが現れた．受付から博士に電話すると，Ashkin，Chandross 両博士が受付までじきじきに出迎えにきてくださった．

当時，われわれが行っていた高分子微粒子１個のレーザー爆蝕について説明すると，興味を示しいろいろと質問をしてくださった．とくに微粒子に１μm 以下の細い穴が開く現象については，今後自己集束などの非線形現象について研究を進めていってはというアドバイスをいただいた．また，博士の実験室を案内していただき，当時博士が取り組まれていたレーザーマニピュレーション法の生物学的応用に関するデータを，ビデオを使って説明していただくことができた．ご高齢にもかかわらず，われわれに実験の説明をされる博士の眼は，少年のような輝きをもっていたことがとても印象的であった．



現在，Ashkin 博士は研究の第一線からは退かれているが，博士の見いだされたレーザーマニピュレーション法は，いまや物理，化学，生物，そして機械工学や化学工学などのさまざまな分野で花開こうとしている．

写真は Ashkin 博士の研究室で撮影したもので，写真右から Chandross 博士，Ashkin 博士，著者(三澤)，喜多村博士，撮影は笹木博士による．

History of Optical Trapping and Manipulation of Small-Neutral Particle, Atoms, and Molecules

A. Ashkin, *Life Fellow, IEEE*

Invited Paper

***Abstract*—This paper reviews the history of optical trapping and manipulation of small-neutral particles, from the time of its origin in 1970 up to the present. As we shall see, the unique characteristics of this technique are having a major impact on the many subfields of physics, chemistry, and biology where small particles play a role.**

equilibrium point in space, with the property that any displacement of a particle away from this point results in a restoring force.

Over the years, these newly found laser trapping and manipu-

I. Introduction

I WILL review the history of optical trapping and manipulation of small neutral particles, with particular emphasis on the origins of the field. This subject, which did not even exist before the advent of lasers, now plays a major role in single particle studies in physics, chemistry, and biology. It was known from physics and the early history of optics that light had linear and angular momentum, and, therefore, could exert radiation pressure and torques on physical objects. These effects were so small, however, that they were not easily detected. To quote J. H. Poynting's presidential address to the British Physical Society in 1905, concerning radiation pressure forces, "A very short experience in attempting to measure these forces is sufficient to make one realize their extreme minuteness—a minuteness which appears to put them beyond consideration in terrestrial affairs …" The study of radiation pressure was considered exciting physics, but not very practical at the turn of the previous century when Nichols and Hull [1] and Lebedev [2] first succeeded in experimentally detecting radiation pressure on macroscopic objects and absorbing gases. The subject essentially dropped into obscurity until the invention of the laser in 1960 [3].

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三四郎も知っていた放射圧

「昼間のうちに，あんな準備をしておいて，夜になって，交通その他の活動が鈍くなるころに，この静かな暗い穴倉で，望遠鏡の中から，あの目玉のようなものをのぞくのです．そうして光線の圧力を試験する．…」

これは夏目漱石が1908年に書いた小説『三四郎』のなかで，理科大学の野々宮先生が研究室を訪ねてきた三四郎に実験の説明をする場面である．夏目漱石は，野々宮先生のモデルとされている門下の寺田寅彦(東京大学教授，地球物理学専攻)から，レベテフとともに放射圧の測定で有名なニコルスの論文〔*Phys. Rev.*, **13**, 307 (1901)； *ibid.*, **17**, 26 (1903)〕の話を聞いて，このような光景を書いたのである．さらに，野々宮先生が実験について，

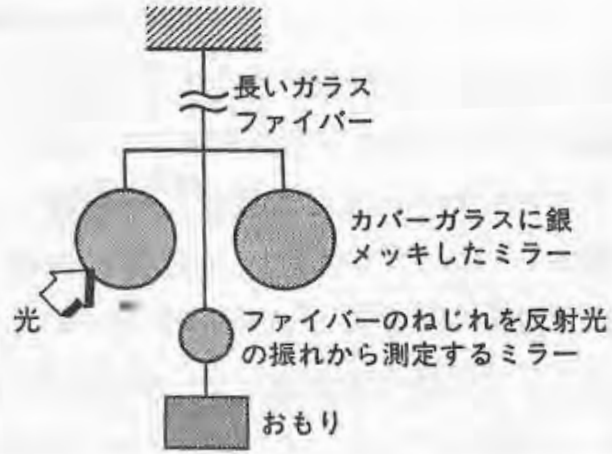
雲母か何かで，十六武蔵ぐらいの大きさの薄い円盤を作って，水晶の糸で釣るして，真空のうちに置いて，この円盤の面へ弧光(アーク)燈の光を直角にあてると，この円盤が光に圧されて動く．

と説明するところがある．これは右図のニコルスの実験装置を実にリアルに描いている．ちなみにニコルスは，この円盤の動きを反射した光の角度から精密に測定して放射圧を計算した．この放射圧の実験に対して，

三四郎は大いに驚いた．驚くとともに光線にどんな圧力があって，その圧力がどんな役に立つんだか，まったく要領を得るに苦しんだ．

と率直な疑問を抱いているが，その答えは1970年のAshkinの論文まで半世紀以上待たなければならなかった．

この三四郎も放射圧を知っていたという話は，ある講演会で東京工業大学の小尾教授に教えていただいたのであるが，残念ながらAshkinのレーザー捕捉の実験がでてくる小説はまだ聞いたことがない．



ACCELERATION AND TRAPPING OF PARTICLES BY RADIATION PRESSURE

A. Ashkin
Bell Telephone Laboratories, Holmdel, New Jersey 07733
(Received 3 December 1969)

Micron-sized particles have been accelerated and trapped in stable optical potential wells using only the force of radiation pressure from a continuous laser. It is hypothesized that similar accelerations and trapping are possible with atoms and molecules using laser light tuned to specific optical transitions. The implications for isotope separation and other applications of physical interest are discussed.

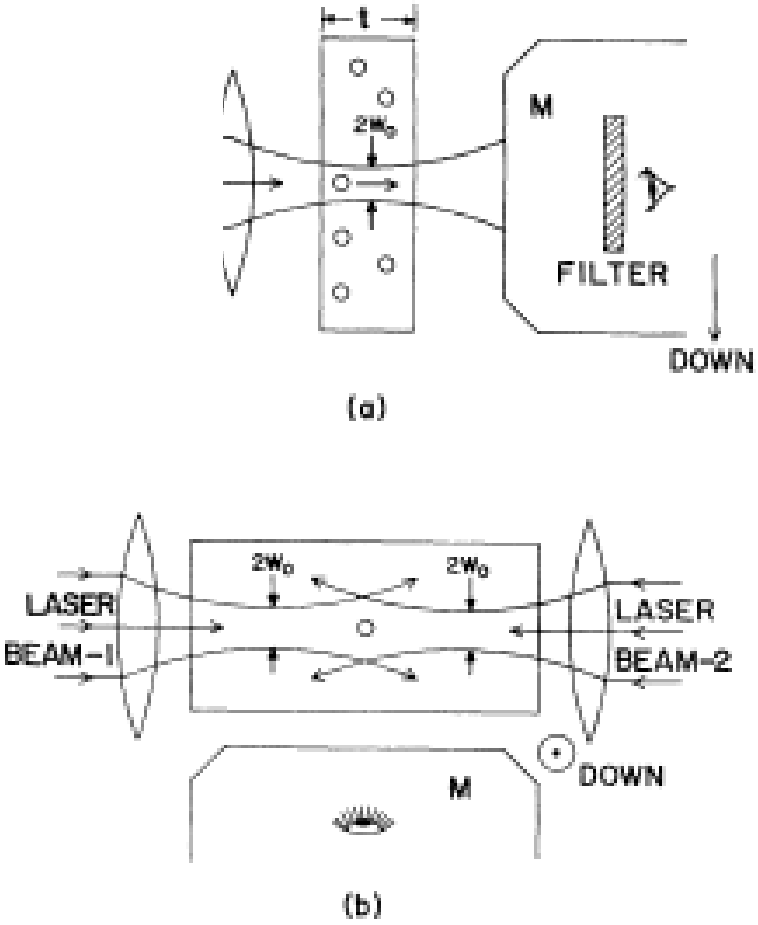


FIG. 1. (a) Geometry of glass cell, $t = 120\ \mu m$, for observing micron particle motions in a focused laser beam with a microscope M. (b) The trapping of a high-index particle in a stable optical well. Note position of the TEM_{00}-mode beam waists.

Optical Levitation by Radiation Pressure

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(Received 14 June 1971; in final form 13 August 1971)

The stable levitation of small transparent glass spheres by the forces of radiation pressure has been demonstrated experimentally in air and vacuum down to pressures ~1 Torr. A single vertically directed focused TEM_{00}-mode cw laser beam of ~250 mW is sufficient to support stably a ~20-μ glass sphere. The restoring forces acting on a particle trapped in an optical potential well were probed optically by a second laser beam. At low pressures, effects arising from residual radiometric forces were seen. Possible applications are mentioned.

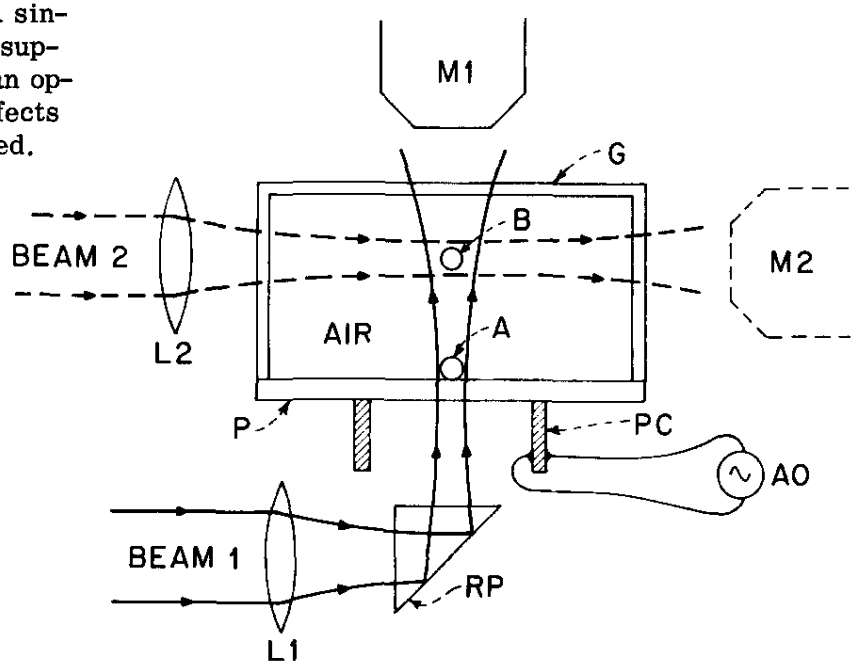

FIG. 1. Levitation apparatus. Particle at A is shaken loose acoustically and lifted to B by TEM_{00}-mode beam 1. TEM_{00}-mode beam 2 is introduced later as a probe beam to study the strength of the trapping forces. L1 and L2 are lenses, P is a glass plate, G is a glass enclosure about 1.5 cm high, RP is a reflecting prism, PC is a piezoelectric ceramic cylinder driven by audio-oscillator AO, and M1 and M2 are microscopes.

Observation of a single-beam gradient force optical trap for dielectric particles

A. Ashkin, J. M. Dziedzic, J. E. Bjorkholm, and Steven Chu

AT&T Bell Laboratories, Holmdel, New Jersey 07733

Received December 23, 1985; accepted March 4, 1986

Optical trapping of dielectric particles by a single-beam gradient force trap was demonstrated for the first reported time. This confirms the concept of negative light pressure due to the gradient force. Trapping was observed over the entire range of particle size from 10 μm to ~25 nm in water. Use of the new trap extends the size range of macroscopic particles accessible to optical trapping and manipulation well into the Rayleigh size regime. Application of this trapping principle to atom trapping is considered.

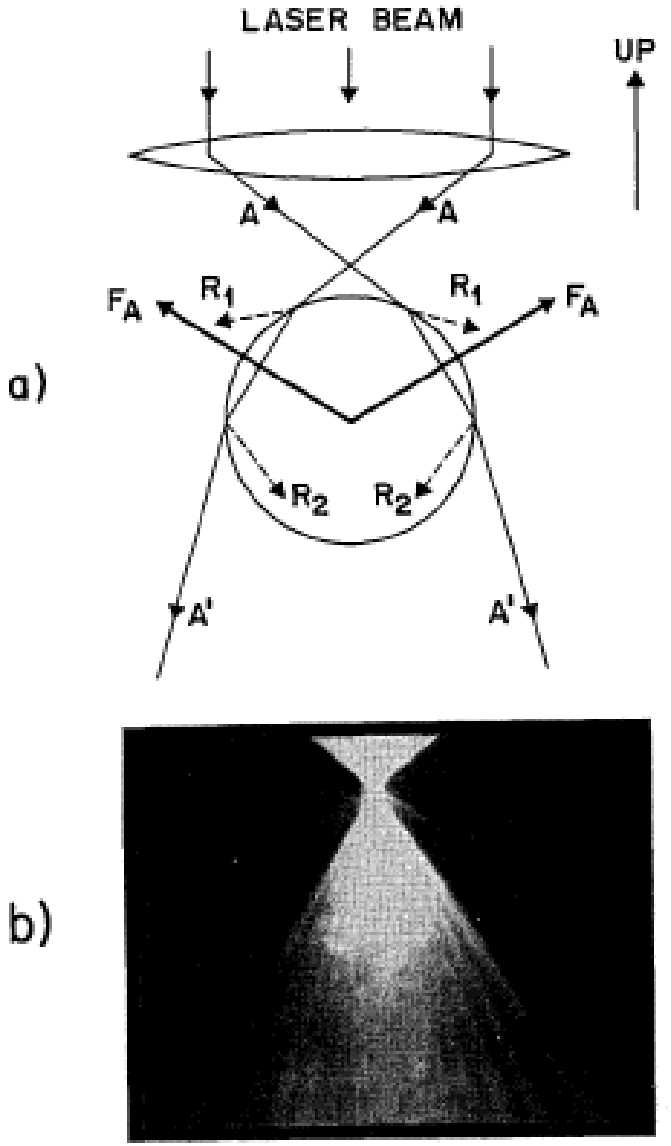

Fig. 1. a) Diagram showing the ray optics of a spherical Mie particle trapped in water by the highly convergent light of a single-beam gradient force trap. b) Photograph, taken in fluorescence, of a 10-μm sphere trapped in water, showing the paths of the incident and scattered light rays.

Optical trapping and manipulation of single cells using infrared laser beams

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† AT&T Bell Laboratories, Murray Hill, New Jersey 07974, USA

Use of optical traps for the manipulation of biological particles was recently proposed, and initial observations of laser trapping of bacteria and viruses with visible argon-laser light were reported[1]. We report here the use of infrared (IR) light to make much improved laser traps with significantly less optical damage to a variety of living cells. Using IR light we have observed the reproduction of *Escherichia coli* within optical traps at power levels sufficient to give manipulation at velocities up to ~500 μm s^{-1}. Reproduction of yeast cells by budding was also achieved in IR traps capable of manipulating individual cells and clumps of cells at velocities of ~100 μm s^{-1}. Damage-free trapping and manipulation of suspensions of red blood cells of humans and of organelles located within individual living cells of spirogyra was also achieved, largely as a result of the reduced absorption of haemoglobin and chlorophyll in the IR. Trapping of many types of small protozoa and manipulation of organelles within protozoa is also possible. The manipulative capabilities of optical techniques were exploited in experiments showing separation of individual bacteria from one sample and their introduction into another sample. Optical orientation of individual bacterial cells in space was also achieved using a pair of laser-beam traps. These new manipulative techniques using IR light are capable of producing large forces under damage-free conditions and improve the prospects for wider use of optical manipulation techniques in microbiology.

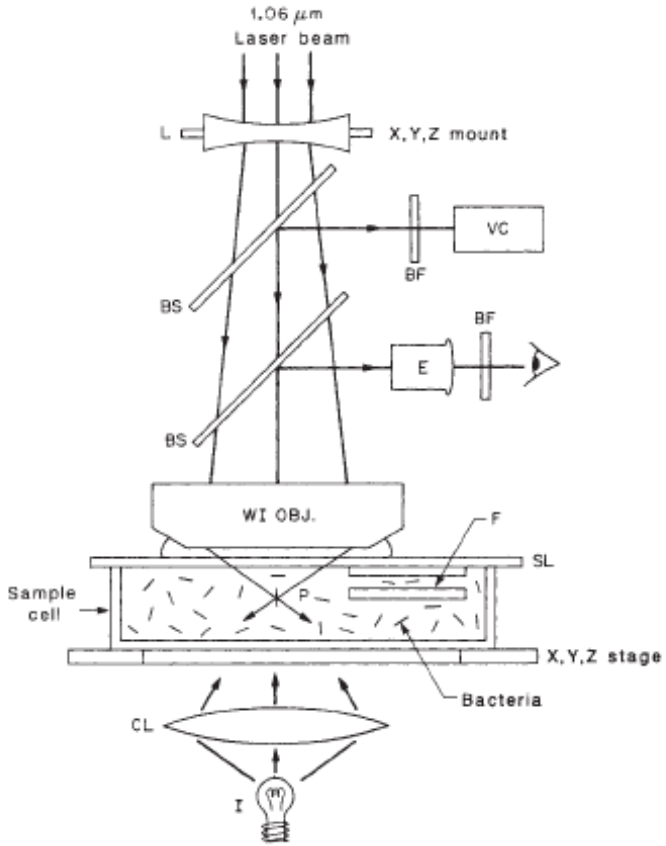

Fig. 1 Combined high-resolution optical microscope and 1.06 μm infra-red laser trap for observing, manipulating and separating bacteria and other organisms.

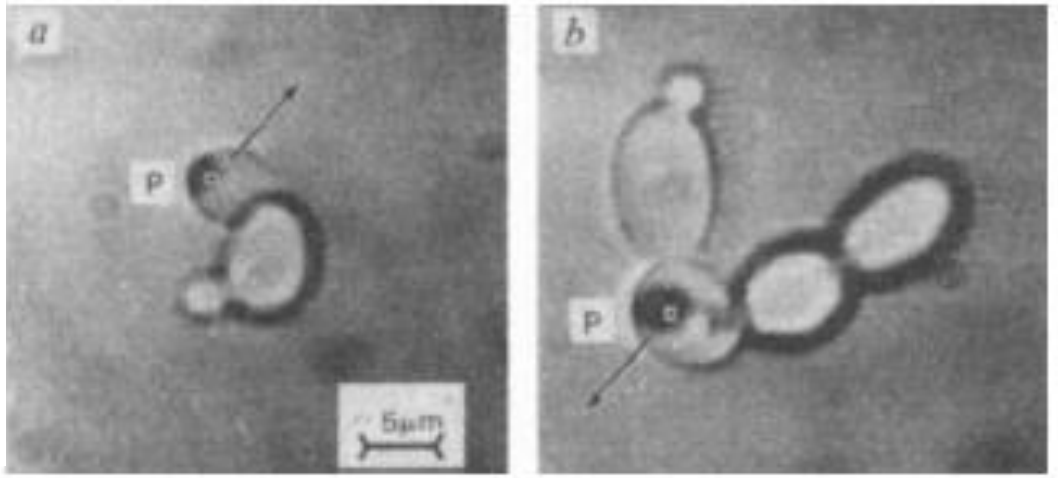

Fig. 2 The division of yeast cells in the IR trap. An original clump of two cells increases to four cells and then to six cells as shown in *a* and *b* after a total elapsed time of about 3 h.

Optical Trapping and Manipulation of Viruses and Bacteria

A. ASHKIN AND J. M. DZIEDZIC

Optical trapping and manipulation of viruses and bacteria by laser radiation pressure were demonstrated with single-beam gradient traps. Individual tobacco mosaic viruses and dense oriented arrays of viruses were trapped in aqueous solution with no apparent damage using ~120 milliwatts of argon laser power. Trapping and manipulation of single live motile bacteria and *Escherichia coli* bacteria were also demonstrated in a high-resolution microscope at powers of a few milliwatts.

size range of 10^5, from ~10 μm down to a few angstroms, which includes both Mie- and Rayleigh-size particles.

The sensitivity of laser trap effectiveness to optical absorption and particle shape is of particular importance for the trapping of biological particles. Absorption can cause an excessive temperature rise or additional thermally generated (radiometric) forces as a result of temperature gradients within a particle (*9*). In general, the smaller the particle size the less the temperature rise and the

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Forces of a single-beam gradient laser trap on a dielectric sphere in the ray optics regime

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ABSTRACT We calculate the forces of single-beam gradient radiation pressure laser traps, also called "optical tweezers," on micron-sized dielectric spheres in the ray optics regime. This serves as a simple model system for describing laser trapping and manipulation of living cells and organelles within cells. The gradient and scattering forces are defined for beams of complex shape in the ray-optics limit. Forces are calculated over the entire cross-section of the sphere using TEM_{00} and TEM^{*}_{01} mode input intensity profiles and spheres of varying index of refraction. Strong uniform traps are possible with force variations less than a factor of 2 over the sphere cross-section. For a laser power of 10 mW and a relative index of refraction of 1.2 we compute trapping forces as high as $\sim 1.2 \times 10^{-6}$ dynes in the weakest (backward) direction of the gradient trap. It is shown that good trapping requires high convergence beams from a high numerical aperture objective. A comparison is given of traps made using bright field or differential interference contrast optics and phase contrast optics.

Proc. Natl. Acad. Sci. USA
Vol. 94, pp. 4853–4860, May 1997
Physics

This contribution is part of the special series of Inaugural Articles by members of the National Academy of Sciences elected on April 30, 1996

Optical trapping and manipulation of neutral particles using lasers

ARTHUR ASHKIN

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Contributed by Arthur Ashkin, March 11, 1997

ABSTRACT **The techniques of optical trapping and manipulation of neutral particles by lasers provide unique means to control the dynamics of small particles. These new experimental methods have played a revolutionary role in areas of the physical and biological sciences. This paper reviews the early developments in the field leading to the demonstration of cooling and trapping of neutral atoms in atomic physics and to the first use of optical tweezers traps in biology. Some further major achievements of these rapidly developing methods also are considered.**

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so-called "

There has been extensive use of optical trapping techniques in the field of microchemistry, which studies the spectroscopy and chemistry of small micron-sized domains. Experiments combining trapping with fluorescence, absorption spectroscopy, photochemistry, and electrochemistry were performed. Polymerization, ablation, and other microfabrication techniques were demonstrated with micrometer samples. Beam-scanning techniques were developed for trapping of micron-sized metal particles, low index particles, and moving of particle arrays in complex patterns. These experiments are described by Masuhara *et al.* (151), summarizing the results of a 5-year Exploratory Research for Advanced Technology project. Bar-Zvi *et al.* (152, 153) have used tweezers to study the physical properties of membranes and

151. Masuhara, H., deSchryver, F. C., Kitamura, N. & Tamai, N. (1994) *Micro-Chemistry-Spectroscopy and Chemistry in Small Domains* (North–Holland, Amsterdam).

History of Optical Trapping and Manipulation of Small-Neutral Particle, Atoms, and Molecules

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Invited Paper

***Abstract*—This paper reviews the history of optical trapping and manipulation of small-neutral particles, from the time of its origin in 1970 up to the present. As we shall see, the unique characteristics of this technique are having a major impact on the many subfields of physics, chemistry, and biology where small particles play a role.**

equilibrium point in space, with the property that any displacement of a particle away from this point results in a restoring force.

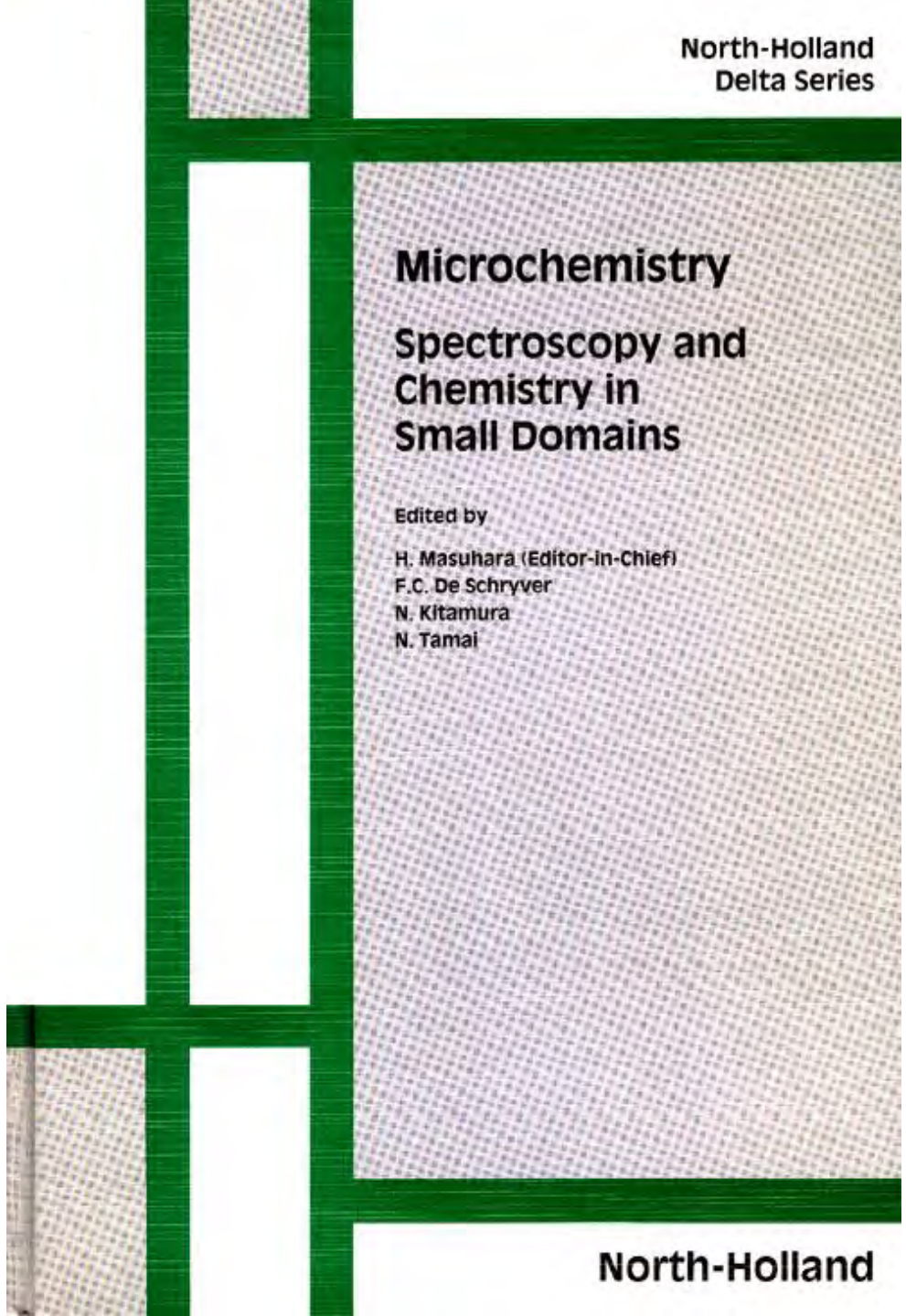
Over the years, these newly found laser trapping and manipu-

Extensive use of optical trapping techniques has been made in the field of microchemistry, which studies the spectroscopy and chemistry of small μm-sized domains. Experiments combining trapping with fluorescence, absorption spectroscopy, photochemistry, and electrochemistry were performed. Polymerization, ablation, and other microfabrication techniques were demonstrated with micron samples. Beam scanning techniques were developed for trapping of μm-sized metal particles, low index particles, and moving of particle arrays in complex patterns. These experiments are by Masuhara *et al.* [183], summarizing the results of a five year ERATO project.

[183] H. Masuhara, F. C. deSchryver, N. Kitamura, and N. Tamai, *Microchemistry-Spectroscopy and Chemistry in Small Domains*: North Holland, 1994.

[184] R. Bar-Ziv, R. Menes, E. Moses, and S. A. Safran, "Local unbinding of pinched membranes," *Phys. Rev. Lett.*, vol. 75, p. 3356, 1995.

[185] R. Bar-Ziv, T. Frisch, and E. Moses, "Entropic expulsion in vesicles," *Phys. Rev. Lett.*, vol. 75, p. 3481, 1995.



IX. The Future

Looking ahead to the early years of the new century, it seems fair to predict that use of optical manipulation techniques will continue to grow at an increasingly rapid pace in the many subfields of physics, chemistry, and biology involving small particles. We are entering an era of increasing emphasis on the small for applications and for basic science. Microtechnology, small machines, small motors, motor molecules, gene sequencing, genetic engineering, and biological computers are already familiar terms. The role of laser tweezers and manipulation in basic sciences has been truly revolutionary. Atomic physics is once again growing in vitality. The cooling of atoms to the lowest temperature yet observed, the achievement of BEC, superfluid behavior of condensates, and atom lasers give new ways of studying quantum effects. The impact of laser technology on the biological sciences may prove to be equally revolutionary.

Our Study
on
Laser Trapping Dynamics and Chemistry in Solution

Trapping and Assembling

J. Phys. Chem. B **1997**, *101*, 5900−5904

Laser-Controlled Association of Poly(*N*-vinylcarbazole) in Organic Solvents: Radiation Pressure Effect of a Focused Near-Infrared Laser Beam

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Received: March 28, 1997; In Final Form: May 22, 1997®

Microassociation of poly(*N*-vinylcarbazole) by a focused near-infrared laser beam (cw-YAG, 1064 nm) was first demonstrated in organic solvents. Investigations were made in cyclohexanone and *N*,*N*-dimethylformamide, where only the radiation force is responsible for formation of the micrometer-sized particle. The following conditions of the formation process were examined for elucidating the mechanism: polymer association as a function of the concentration (range 3.5−6.8 wt %), the relation between the trapping laser power and the time of condensation, and the influences of the temperature and the solvent properties upon the formation process.

J. Phys. Chem. B **1999**, *103*, 1660−1663

Photon Pressure-Induced Association of Nanometer-Sized Polymer Chains in Solution

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Received: September 11, 1998; In Final Form: January 4, 1999

The photon pressure effect induced by a focused infrared laser beam upon two kinds of water-soluble carbazolyl-containing copolymers in aqueous solution is shown to result in the formation of single microparticles. The minimum size of the polymer chains at which the photon pressure overcomes the Brownian motion and the electrostatic repulsion is determined to be 10−20 nm. The rate of particle formation and the final particle size are shorter and larger, respectively, as the carbazolyl content of the polymer is higher. This indicates that the polarizability plays an important role in the photon pressure-induced association process.

J. Phys. Chem. B **1998,** *102,* 1896−1901

Chemical and Optical Mechanism of Microparticle Formation of Poly(*N*-vinylcarbazole) in *N*,*N*-Dimethylformamide by Photon Pressure of a Focused Near-Infrared Laser Beam

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Received: September 26, 1997; In Final Form: January 5, 1998

We focus our attention to the laser-controlled association process of poly(*N*-vinylcarbazole) by "pure" photon pressure effect. A few wt % *N*,*N*-dimethylformamide solution of the polymer was irradiated with a focused 1064 nm laser beam of sub wattage power, and a resultant condensation (microparticle formation) at a focal point of the microscope was initially probed by backscattering of He−Ne laser and later observed by transmission image of the microscope. To understand in detail the behavior of the polymer induced by the photon force, the relation between laser power and particle diameter, as well as repeated irradiation effect, were investigated. The laser-controlled association was successfully demonstrated even for short polymer chains of about 20 mean degrees of polymerization. Fluorescence spectra and their rise and decay curves were measured microspectroscopically for a formed microparticle. The results are different from those of the polymer in solution, suggesting the association structure is characteristic of photon pressure effect.

J. Phys. Chem. B **1998,** *102,* 8440−8451

Transmission and Confocal Fluorescence Microscopy and Time-Resolved Fluorescence Spectroscopy Combined with a Laser Trap: Investigation of Optically Trapped Block Copolymer Micelles

Thomas Gensch,[†] Johan Hofkens,[†] Jan van Stam,[†] Herman Faes,[†] Serge Creutz,[‡] Kenji Tsuda,[†] Robert Jérôme,[‡] Hiroshi Masuhara,[§] and Frans C. De Schryver*,[†]

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J. Phys. Chem. B, Vol. 102, No. 40, 1998

Laser-Controlled Assembling of Repulsive Unimolecular Micelles in Aqueous Solution

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Received: June 11, 1998; In Final Form: August 12, 1998

Laser manipulation techniques were applied to the control of molecular assembling in D_2O. An amphiphilic random copolymer, which has hydrophilic and hydrophobic segments in a single chain and forms a monopolymer micelle through self-organization, is demonstrated to receive photon pressure of a focused near-infrared laser beam. Upon prolonged irradiation with the beam, a single microparticle is formed at a focal point, and it dissolves very quickly after switching off the laser beam. The behavior indicates that electrostatic repulsion between micelles is overcome by photon pressure. The assembling processes were examined as a function of irradiation time and polymer concentration and are discussed.

2005, *109*, 6917−6921
Published on Web 03/17/2005

Assembling and Orientation of Polyfluorenes in Solution Controlled by a Focused Near-Infrared Laser Beam

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Received: December 26, 2004; In Final Form: February 21, 2005

Ordered fibril- and particle-like assemblies of poly(2,7-(9,9-bis(2-ethylhexyl)fluorene)) can be formed by photon force of a focused near-infrared laser beam during the drying process of its tetrahydrofuran solution on a glass substrate. These formations have been achieved controllably by combining laser irradiation with convection in the cast solution; that is, when viscous drag of the solution in the convection is stronger than the photon force, the fibril-like assemblies can be formed. Molecular orientation in the assemblies differs from that in self-assembled fibril-like structures, and maybe it can be controlled by the polarization direction of the focused laser beam. We have demonstrated that the length and width of the assemblies can be controlled by the irradiation time, the laser power, the concentration of the solution, and the convection rate in the solution. On the other hand, when the viscous drag of the solution in the convection is weak compared to the photon force, particle-like assemblies in which molecular orientation is controlled by polarization direction are formed.

PHYSICAL REVIEW E 70, 061406 (2004)

Reversible assembly of gold nanoparticles confined in an optical microcage

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(Received 28 July 2004; published 13 December 2004)

As optical trapping by a focused laser beam is applied for nanoparticles, multiple particles are grasped in the focal spot and make an assembly. Gold nanoparticles confined in such a submicrometer optical cage show a characteristic extinction spectrum depending on laser power. The spectral change can be induced reversibly and repeatedly by tuning the laser power, demonstrating that the assembly of gold nanoparticles can be controlled by the gradient force. This is attributed to the soft confinement of nanoparticles dressed in electrostatic potential barriers.

Thiacarbocyanine dye J-aggregation in optical trapping potential

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ABSTRACT

Optical assembly of J-aggregates has been investigated by focusing a CW Nd^{3+}:YAG laser (λ=1064nm) in THIATS solution. Spectroscopic property of THIATS dye was analyzed by using a confocal fluorescence microscope. As a result, we found that fluorescence spectra, or electric structure, of the THIATS J-aggregates change following the increase of concentration. These THIATS J-aggregates were trapped and gathered at the focal point by focusing NIR laser beam into THIATS solution. Then fluorescence from the focal spot was concurrently detected with the same trapping laser beam, which is due to NIR two-photon absorption because of its extremely high intensity. Fluorescence spectrum at the focal spot changed similarly to the increase of concentration. This result could be explained by the transformation into J-aggregates with higher polarizability in the focal spot of NIR laser. Furthermore, we have succeeded in depositing J-aggregates, whose structure is selected, in the vicinity of the focal spot by optical trapping.

Keywords: J-aggregates, optical trapping, self-assembly, 3,3'-bis-[3-sulfopropyl]-5,5'-dichloro-9-ethyl thiacarbocyanine, supramolecular aggregates, nanotechnology, nanoscience, photon pressure, two-photon spect[1]scopy, confocal fluorescence image, evaporation process

J. Phys. Chem. B **2006,** *110,* 17906−17911

Two-Photon Fluorescence Spectroscopy of Individually Trapped Pseudoisocyanine J-Aggregates in Aqueous Solution

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Received: May 23, 2006; In Final Form: July 24, 2006

We have investigated a pseudoisocyanine dye aqueous solution including nanometer-sized J-aggregates by combining optical trapping and two-photon fluorescence spectroscopy. By focusing an intense near-infrared laser into an 8×10^{-3} M solution, the intense fluorescence from J-aggregates for a few to tens of seconds is observed intermittently, indicating that individual J-aggregates are trapped in and diffuse out from a focal spot. The peak position and full width at half-maximum of the J-band are different from each other. By measuring 171 J-aggregates, it was found that J-aggregates can be classified largely into two groups. The existence of two kinds of groups of J-aggregates could be attributed to the difference in the nucleation process, which is affected by the substrate. J-aggregates possessing a J-band of a narrower bandwidth in a shorter wavelength region are trapped for a longer period of time, indicating that highly ordered J-aggregates are trapped for a longer period of time because of their high polarizability.

THE JOURNAL OF PHYSICAL CHEMISTRY C LETTERS

2007, *111*, 18457−18460
Published on Web 11/22/2007

Laser-Induced Self-Assembly of Pseudoisocyanine J-Aggregates

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Received: October 12, 2007; In Final Form: November 13, 2007

This letter describes that the appearance and growth of J-aggregate are clearly accelerated by focusing a near-infrared laser beam into an aqueous solution of psudoisocyanine while gradually increasing the concentration by solvent evaporation. The J-aggregate formed in the laser focus shows a narrower fluorescence bandwidth compared to those obtained by simple concentration increase. This indicates that the growth of the highly ordered J-aggregates is preferentially enhanced in the laser focus, which is the first demonstration showing laser-induced self-assembly of psudoisocyanine J-aggregates. Furthermore, we discuss the mechanism of the laser-induced self-assembly on the basis of thermodynamic equilibrium between the in and out of optical trapping potential produced by the focused laser beam.

Selective Trapping

THE JOURNAL OF PHYSICAL CHEMISTRY B LETTERS

2006, *110*, 21399−21402
Published on Web 10/11/2006

Selective Optical Trapping and Deposition of Polymer and Aromatic Molecules from Binary Mixed Solution

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Received: August 30, 2006; In Final Form: September 27, 2006

We have demonstrated size-selective optical trapping and deposition of polymer and aromatic molecules from binary mixed solution. As a near-infrared laser beam is tightly focused in polystyrene and perylene mixed solution and dropped on a glass substrate, a molecular assembly is deposited at the laser focus and fixed on the substrate. The fluorescence spectrum of the deposited microassembly depends on the laser power; perylene monomer fluorescence is dominant in the case of high laser power, whereas excimer emission of perylene crystal is observed in the case of low laser power. This suggests that polystyrene molecules are preferentially deposited by focusing a higher laser power so that the ratio of polystyrene and perylene in the assembly can be controlled by laser power. This mechanism can be explained in view of the molecular size selectivity in optical trapping.

Trapping and Patterning

Appl. Phys. Lett., Vol. 78, No. 17, 23 April 2001

Optical patterning and photochemical fixation of polymer nanoparticles on glass substrates

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(Received 7 December 2000; accepted for publication 21 February 2001)

A method for fixing patterned nanoparticles onto a substrate was developed by combining photopolymerization with a laser manipulation technique. Nanoparticles were dispersed in ethylene glycol containing monomer, crosslinker, and photoinitiator, and gathered at the focal point of a trapping laser beam (1064 nm) just on a glass substrate. Local photopolymerization within and around the nanoparticles assembly was induced by additional irradiation of a pulsed-laser beam (355 nm), resulting in generation of polyacrylamide containing nanoparticles. The polymerized assembly was evaluated by atomic force microscope observation. By scanning both trapping and excitation laser beams, patterned nanoparticles could also be fixed on a glass substrate. [DOI: 10.1063/1.1366646]

Appl. Phys. Lett., Vol. 80, No. 3, 21 January 2002

Laser manipulation and fixation of single gold nanoparticles in solution at room temperature

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(Received 2 August 2001; accepted for publication 14 November 2001)

A method to fix single gold nanoparticles on glass substrate was demonstrated in solution at room temperature by utilizing focused intense laser beams. A single gold nanoparticle of 80 nm was optically trapped and manipulated to a surface of a glass substrate, and then fixed on it by irradiation with ultraviolet (UV) laser light. Suitable laser fluence range for the fixation was determined to be 32–60 mJ/cm^2, above which the individual nanoparticles were fragmented to several smaller fragments of 10 to 40 nm. The fixation mechanism is discussed in view of pulsed-laser-induced transient temperature elevation. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1432753]

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Vol. 46, No. 1, 2007, pp. 449–454

Laser Deposition of Polymer Micro- and Nanoassembly from Solution Using Focused Near-Infrared Laser Beam

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(Received April 10, 2006; accepted September 30, 2006; published online January 10, 2007)

We have demonstrated the laser deposition of polymer micro- and nanoassemblies from a solution onto a glass substrate. The size and shape of the deposited dot-like assembly can be controlled by the laser power (P) and the concentration of the solution (C). For an example, a nanoassembly of a π-conjugated polymer, whose width and height are 280 and 23 nm, respectively, is deposited at the conditions of $C = 1.0 \times 10^{-5}$ mg/ml and $P = 700$ mW. This laser deposition can be attributed to the optical trapping and the surface deformation of the solution layer using a focused laser beam. It is also demonstrated that the molecular orientation in the assembly can be aligned in the direction of the laser polarization. The present laser deposition is applicable to the micropatterning of various polymers dissolved in an organic solvent.
[DOI: 10.1143/JJAP.46.449]

KEYWORDS: optical trapping, laser deposition, laser microfabrication, thermocapillary, π-conjugated polymer

dx.doi.org/10.1021/ja200737j | J. Am. Chem. Soc. 2011, 133, 14472–14475

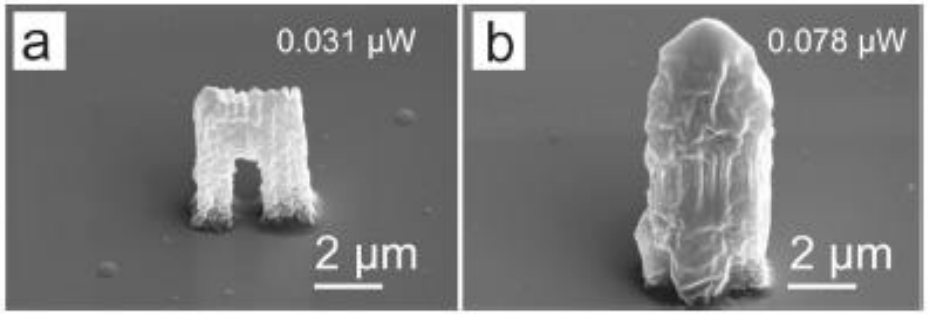

Figure 4. (a) Microstructure resembling the "Arch of Triumph" produced by simultaneous irradiation with UV and NIR lasers. (b) Microstructure produced by UV irradiation only. The two structures (a and b) were produced via the same procedure with the same blueprint. Applied UV powers are shown in the figures.

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Confinement of Photopolymerization and Solidification with Radiation Pressure

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APPLIED PHYSICS LETTERS 91, 041102 (2007)

Laser microfixation of highly ordered *J* aggregates on a glass substrate

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(Received 21 April 2007; accepted 25 June 2007; published online 23 July 2007)

The authors have developed a laser microfixation technique of *J* aggregates from pseudoisocyanine solution onto a glass substrate. By focusing a near-infrared (NIR) laser beam into the solution near the glass substrate, conglomerate *J* aggregates are deposited at the focal point and fixed on the substrate. Fixed *J* aggregates show a narrower fluorescence band than the deposited one without laser focusing, indicating that highly ordered *J* aggregates are selectively confined in the laser focus due to optical trapping. Furthermore, they have demonstrated that the orientation of the transition dipole in fixed *J* aggregates is controlled by the polarization direction of the NIR laser beam.
 [DOI: 10.1063/1.2759468]

Trapping-induced Morphological Change and Phase Transition

A Single Droplet Formation from Swelled Micelles by Radiation Pressure of a Focused Infrared Laser Beam

J. Am. Chem. Soc. **1996**, *118*, 11968−11969

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Received May 22, 1996
Revised Manuscript Received August 19, 1996

Radiation pressure due to photon momentum change was prospected by Newton, theoretically proved by Maxwell, and experimentally confirmed by Lebedev.[1] Based on the radiation pressure, a micrometer particle in solution can be trapped by a laser beam, which was demonstrated for the first time by Ashkin.[2] The Brownian motion of microparticles is completely suppressed without any contact and destruction, hence the trapping method is very useful to treat microparticles in solution. We have developed this method further to manipulate a single or plural microparticle(s) freely in three-dimensional space by introducing a computer-controlled pair of galvano mirrors; scanning laser micromanipulation.[3] Now, chemical applications are conducted by combining this method with fluorescence spectroscopy, photochemical reaction, laser ablation, and electrochemistry using a microelectrode.[4] It is also known that radiation pressure is exerted upon Rayleigh particles that are smaller than the wavelength. Therefore, spectroscopy and chemistry of a single molecular assembly, quantum particle, and so on in solution is expected.

Mol. Cryst. Liq. Cryst., 1996, Vol. 283, pp. 165–172

LASER INDUCED PHASE TRANSITION IN AQUEOUS SOLUTIONS OF HYDROPHOBICALLY MODIFIED POLY(N-ISOPROPYLACRYLAMIDE).

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Abstract : Phase transitions of amphiphilic copolymer solutions can be achieved by irradiating aqueous solutions of this polymer with an IR-laser beam. As a result, a single microparticle up to 15 μm is formed, whereas thermal heating of the solution results in submicrometer particles. Both the photo-thermal effect (due to absorption of 1064 nm by the H_2O overtone) and the radiation force cause the unusual large particle formation (up to 15 μm). Furthermore, the present study shows that the conformational distribution and associations of polymers in solution can be controlled with the radiation force of a focused laser beam.

Langmuir **1997**, *13*, 414−419

Molecular Assembling by the Radiation Pressure of a Focused Laser Beam: Poly(*N*-isopropylacrylamide) in Aqueous Solution

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Received June 25, 1996. In Final Form: November 5, 1996[⊗]

Phase transitions in aqueous solutions of poly(*N*-isopropylacrylamide) (PNIPAM) with a molecular weight ($\bar{M}_w$) of 63 000 were achieved by irradiating the solutions (0.2−3.6 wt %) with an IR laser beam (1064 nm) through an optical microscope. First, a microparticle with the size of the focused laser beam was formed (≃1.5 μm). This microparticle continuously grew and after prolonged irradiation (up to 10 min), a microparticle with a maximum size of 25 μm was obtained. Upon further irradiation, the microparticle became unstable and finally disappeared. The importance of the optical alignment of the microscope/laser system is discussed. Particle formation was also found in D_2O solutions of PNIPAM. These experimental results indicate that, besides a photothermal effect (heating up of the solution due to absorption of water at 1064 nm), there is influence of the "radiation force" upon particle formation and conformation properties of the polymer. The observations mentioned above are discussed in connection with the theory of the single beam gradient force optical trap for dielectric particles.

Appl. Phys. Lett. 71 (15), 13 October 1997

Manipulation of liquid crystal textures with a focused near infrared laser beam

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(Received 19 May 1997; accepted for publication 15 August 1997)

Optical manipulation of disclinations and defects in liquid crystal films was demonstrated and discussed in terms of mass transfer induced by radiation pressure and of molecular rotation under the optical electric field. Orientation of liquid crystal molecules was controlled by changing the polarization direction of a focused cw laser beam. A disclination line could be deformed by moving the focal spot, just like drawing a bow. A point defect followed the laser beam so that it could be freely transported in the film. When two disclination points were optically manipulated to become fused, the defects disappeared immediately and did not return after switching off the laser. [S0003-6951(97)01041-3]

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2005, *109*, 6917−6921
Published on Web 03/17/2005

Assembling and Orientation of Polyfluorenes in Solution Controlled by a Focused Near-Infrared Laser Beam

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Received: December 26, 2004; In Final Form: February 21, 2005

Ordered fibril- and particle-like assemblies of poly(2,7-(9,9-bis(2-ethylhexyl)fluorene)) can be formed by photon force of a focused near-infrared laser beam during the drying process of its tetrahydrofuran solution on a glass substrate. These formations have been achieved controllably by combining laser irradiation with convection in the cast solution; that is, when viscous drag of the solution in the convection is stronger than the photon force, the fibril-like assemblies can be formed. Molecular orientation in the assemblies differs from that in self-assembled fibril-like structures, and maybe it can be controlled by the polarization direction of the focused laser beam. We have demonstrated that the length and width of the assemblies can be controlled by the irradiation time, the laser power, the concentration of the solution, and the convection rate in the solution. On the other hand, when the viscous drag of the solution in the convection is weak compared to the photon force, particle-like assemblies in which molecular orientation is controlled by polarization direction are formed.

Langmuir **2007**, *23*, 6725−6729

Effects of Optical Trapping and Liquid Surface Deformation on the Laser Microdeposition of a Polymer Assembly in Solution

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Received November 15, 2006. In Final Form: March 21, 2007

A polymer microassembly is formed by focusing a near-infrared (NIR) laser beam in a thin film of a polymer solution. We have investigated the mechanism of laser microdeposition of a polyfluorene assembly by measuring the surface deformation of the solution film and the morphology of the deposited assembly. It is clearly observed that a rupture is formed at the laser focus in the solution film by using laser interferometric imaging. The time necessary for the rupture formation and the volume of the deposited microassembly are analyzed as a function of laser power. Experimental results suggest that the solution surface deformation induced by local laser heating and optical trapping effects determined the volume of the laser microdeposition. By combining this method with multiple optical trapping, a polymer microassembly with a polygonal morphology is formed on the glass substrate.

Photon Force-Induced Phase Transition Dynamics of Single Hydrogel Nanoparticles in Water

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Kitauoya-Nishimachi, Nara 630-8506, Japan

Dedicated to Late Professor Jerzy Prochorow

We investigated laser-induced phase transition dynamics of μm sized single poly(*N*-isopropylacrylamide) hydrogel particles in water. Single poly(*N*--isopropylacrylamide) gel particles labeled with a polarity-sensitive fluorescent probe monomer were optically trapped by a focused laser beam and its fluorescence dynamics was analyzed. The fluorescence intensity of trapped single gel particles was increased with the trapping laser power, while the fluorescence peak wavelength was not changed. Temperature-induced changes of fluorescence properties of the single particle were confirmed to be similar to those of the bulk solution. These behaviors are well interpreted by considering that the fluorescence intensity and fluorescence peak reflect local interactions between the fluorescent probe and attaching (bound) water molecules and effective polarity determined by (free) water content in the particle, respectively. A change in the fluorescence peak wavelength after laser trapping was followed and its blue-shift was confirmed to occur within a few hundreds seconds, indicating that a single gel particle gradually attains to a globular state on this timescale, expelling initially free water molecules and then bound ones.

PACS numbers: 87.80.Cc, 33.50.Dq, 61.25.Hq, 87.64.–t

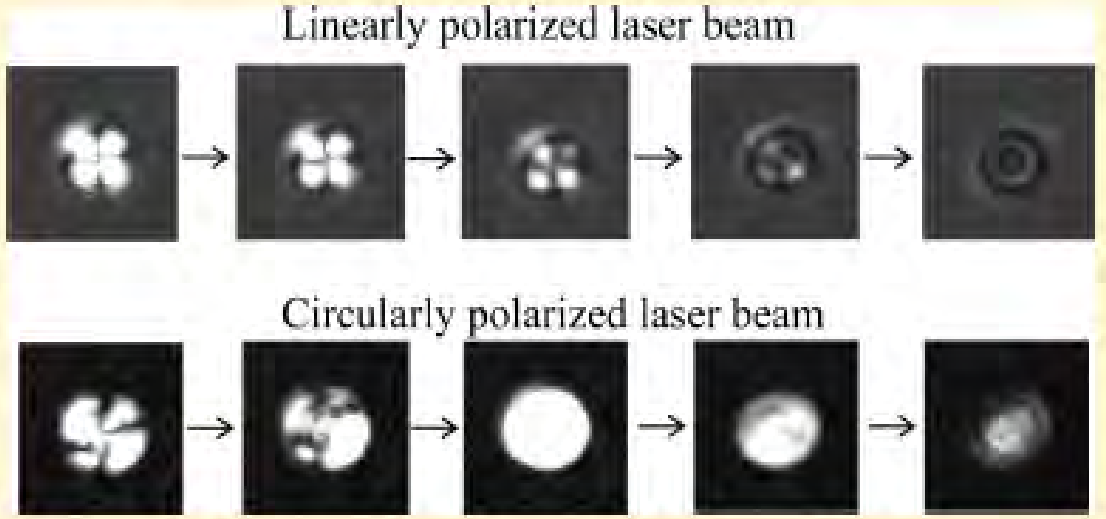
J. Phys. Chem. B 2013, 117, 4536−4540

Polarization and Droplet Size Effects in the Laser-Trapping-Induced Reconfiguration in Individual Nematic Liquid Crystal Microdroplets

Anwar Usman,* Wei-Yi Chiang, Takayuki Uwada, and Hiroshi Masuhara*

Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, Hsinchu 30010, Taiwan

ABSTRACT: We experimentally demonstrate reordering throughout the inside of an individual bipolar nematic liquid-crystalline microdroplet optically trapped by a highly focused laser beam, when the laser powers are above a definite threshold. The threshold depends on the droplet size and laser polarization. A physical interpretation of the results is presented by considering the nonlocal orientations of the nematic liquid-crystal molecules in the droplets with the dimensions on the order of the focal spot diameter or larger. On the basis of the finite size approximation, we show that the dependence of threshold power on the droplet size is calculated to be in qualitative agreement with the experimental data.



Our Study
on
Laser Trapping Dynamics and Chemistry at Interface

Chemistry Letters Vol.36, No.12 (2007)

Crystallization of Glycine by Photon Pressure of a Focused CW Laser Beam

Teruki Sugiyama,* Takuji Adachi, and Hiroshi Masuhara*
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(Received September 21, 2007; CL-071047; E-mail: sugiyama@hlsrf.or.jp)

An intense CW-YVO_4 laser beam of 1064 nm was focused into a supersaturated D_2O solution of glycine, leading to its crystallization. At the focal point on an air–solution interface, a small single glycine crystal was trapped and grew quickly. The crystallization mechanism is considered in view of gathering and organization of large solute clusters of glycine by photon pressure.

Chemistry Letters Vol.38, No.5 (2009)

Crystal Growth of Glycine Controlled by a Focused CW Near-infrared Laser Beam

Teruki Sugiyama,*[1] Takuji Adachi,[1] and Hiroshi Masuhara*[1,2]

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[2]*Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, Hsinchu 30010, Taiwan*

(Received January 29, 2009; CL-090108; E-mail: sugiyama@ms.naist.jp)

Crystal growth of glycine was accelerated just by focusing a CW 1064-nm laser beam at a position adjacent to a spontaneously generated glycine crystal in D_2O. Its rate depended on the distance between the crystal and the focal spot. Interesting crystal growth and dissolution were found, which is considered to be due to Ostwald ripening.

CRYSTAL GROWTH & DESIGN
Communication
2010, Vol. 10
4686–4688

DOI: 10.1021/cg100830x

Crystallization in Unsaturated Glycine/D_2O Solution Achieved by Irradiating a Focused Continuous Wave Near Infrared Laser

Thitiporn Rungsimanon,[†] Ken-ichi Yuyama,[†] Teruki Sugiyama,*[,†] and Hiroshi Masuhara*[,†,‡]

[†]*Graduate School of Materials Science, Nara Institute of Science and Technology, Ikoma 630-0192, Japan, and* [‡]*Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, Hsinchu 30010, Taiwan*

Received June 22, 2010; Revised Manuscript Received September 12, 2010

ABSTRACT: The crystallization of glycine in unsaturated solution is made possible by laser trapping of its molecular clusters due to photon pressure of a focused continuous wave near-infrared laser beam. Always one single crystal is spatiotemporally formed at a focal spot, and then it undergoes dissolution, eventually leading to repetitive crystallization and dissolution. The polymorph characterization of the crystal formed in unsaturated solution confirmed the γ-form, which is not obtainable by conventional crystallization methods. The preparation probability of the γ-form compared to the α-form is much higher than that in the supersaturated solution.

THE JOURNAL OF PHYSICAL CHEMISTRY Letters

pubs.acs.org/JPCL

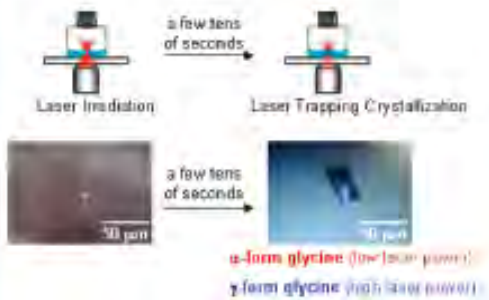
Control of Crystal Polymorph of Glycine by Photon Pressure of a Focused Continuous Wave Near-Infrared Laser Beam

Thitiporn Rungsimanon,[†] Ken-ichi Yuyama,[†] Teruki Sugiyama,*[,†] Hiroshi Masuhara,*[,†,‡] Norimitsu Tohnai,[§] and Mikiji Miyata[§]

[†]Graduate School of Materials Science, Nara Institute of Science and Technology, 8916-5 Takayama, Ikoma, Nara 630-0192, Japan, [‡]Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, Hsinchu 30010, Taiwan, and [§]Department of Material and Life Science, Graduate School of Engineering, Osaka University, Suita 565-0871, Japan

ABSTRACT Crystal polymorph of glycine is controlled by tuning the power of a linearly polarized continuous wave 1064-nm laser beam. Upon focusing the beam to the air/solution interface of a supersaturated glycine/D_2O solution, its single crystal is spatiotemporally formed at a focal spot within a few seconds to a few tens of seconds. Fourier transform infrared measurement and single-crystal X-ray crystallographic analysis of the fabricated single crystal reveal that two polymorphs of α- and γ-forms are prepared depending on the laser power. The probability of γ-form preparation, which is not available under ambient conditions, arises up to 50% at 1.3 W laser power after an objective lens. The mechanism of the polymorph control is discussed in view of both photon pressure and local temperature elevation due to laser irradiation at the focal spot.

SECTION Nanoparticles and Nanostructures



J. Phys. Chem. Lett. 2010, 1, 599–603

THE JOURNAL OF PHYSICAL CHEMISTRY Letters

pubs.acs.org/JPCL

Millimeter-Scale Dense Liquid Droplet Formation and Crystallization in Glycine Solution Induced by Photon Pressure

Ken-ichi Yuyama,[†] Teruki Sugiyama,*[,†] and Hiroshi Masuhara*[,†,‡]

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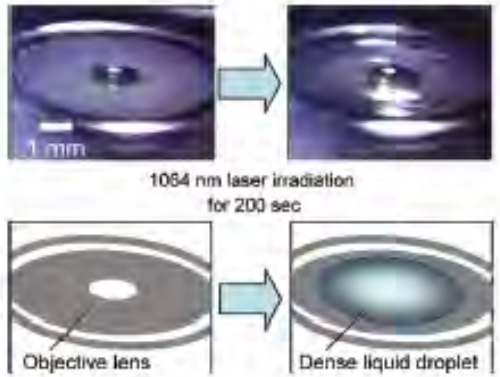
ABSTRACT A millimeter-scale dense liquid droplet of glycine is prepared by focusing a CW near-infrared laser beam at the glass/solution interface of a thin film of its supersaturated heavy water solution. The formation process is investigated by direct observation with CCD and by measuring temporal change of the surface height with a displacement meter. The droplet becomes much larger than a focal spot size, a few mm width and ~150 μm height, and observable with the naked eye. Interestingly, the droplet remains for a few tens of seconds even after switching off the laser beam. Whereas the droplet is kept during laser irradiation, the crystallization is immediately attained by shifting the laser beam to the air/droplet surface. It is considered that the droplet is possibly the early stage of the multistep crystallization process and plays an important role in photon pressure-induced crystallization of glycine.

SECTION Nanoparticles and Nanostructures



Phys. Chem. C 2012, 116, 6809−6816

THE JOURNAL OF PHYSICAL CHEMISTRY C

Article

pubs.acs.org/JPCC

Formation, Dissolution, and Transfer Dynamics of a Millimeter-Scale Thin Liquid Droplet in Glycine Solution by Laser Trapping

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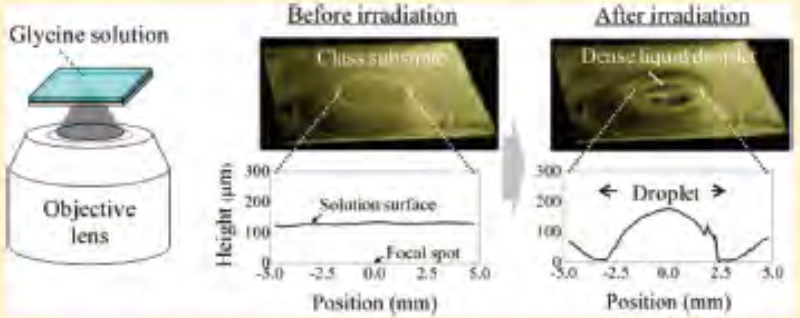
Supporting Information

ABSTRACT: The formation, dissolution, and transfer of a millimeter-scale dense liquid droplet are demonstrated by focusing a CW near-infrared laser beam into a thin film of glycine solution in heavy water. The entire process is investigated by directly monitoring the temporal change in the two-dimensional surface profile using a laser displacement meter. Upon laser irradiation, the surface depression is initially induced by laser heating, followed by the formation of the shallow convex-shaped droplet around the focal spot, in which the droplet is always in contact with the surrounding solution through the ultrathin solution layer. After the laser is switched off, the dissolution occurs through the recovery from the ultrathin layer toward the original solution film. When the laser is set to the outside of the droplet, the solution depression is similarly induced, and subsequently the droplet starts moving toward the focal spot. These processes are summarized and discussed in view of laser-induced effects of concentration increase and temperature elevation.



Cryst. Growth Des. 2012, 12, 2427−2434

CRYSTAL GROWTH & DESIGN

Article

pubs.acs.org/crystal

Selective Fabrication of α- and γ-Polymorphs of Glycine by Intense Polarized Continuous Wave Laser Beams

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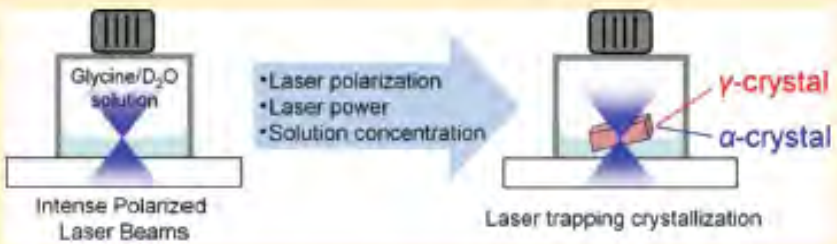
[‡]Graduate School of Materials Science, Nara Institute of Science and Technology, Ikoma 630-0192, Japan

[§]Instrument Technology Research Center, National Applied Research Laboratories, Hsinchu 30076, Taiwan

ABSTRACT: The polymorph control of glycine in D_2O solution is successfully demonstrated by a laser trapping technique using a linearly or circularly polarized CW near-infrared laser beam. Focusing each laser beam into an air/solution interface of the solution always generates the stable crystal polymorph of either α- or γ-form at the focal spot. The formation probability of each polymorph strongly depends on various experimental conditions of laser polarization, power, and solution concentration. For the supersaturated and saturated solutions, circularly polarized laser irradiation enhances γ-crystal formation, while for the unsaturated solution the laser polarization effect becomes prominent and linearly polarized laser light at a specific power provides the maximum γ-crystal probability of 90%. The present polymorphism is achieved by laser-induced effects such as concentration increase, temperature elevation, and molecular rearrangement, whose mechanism is discussed in view of laser polarization dependence of these effects.



J. Phys. Chem. Lett. 2013, 4, 2436−2440

THE JOURNAL OF PHYSICAL CHEMISTRY Letters

Letter

pubs.acs.org/JPCL

Laser Trapping and Crystallization Dynamics of L-Phenylalanine at Solution Surface

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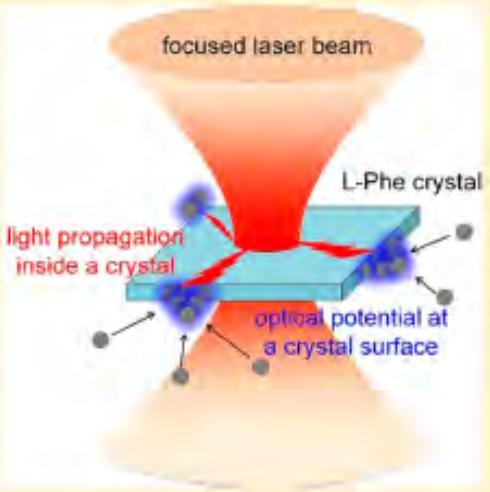
[†]Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, Hsinchu 30010, Taiwan

[‡]Instrument Technology Research Center, National Applied Research Laboratories, Hsinchu 30076, Taiwan

Supporting Information

ABSTRACT: We present laser trapping behavior of L-phenylalanine (L-Phe) at a surface of its unsaturated aqueous solution by a focused continuous-wave (CW) near-infrared (NIR) laser beam. Upon the irradiation into the solution surface, laser trapping of the liquid-like clusters is induced concurrently with local laser heating, forming an anhydrous plate-like crystal at the focal spot. The following laser irradiation into a central part of the plate-like crystal leads to laser trapping at the crystal surface not only for L-Phe molecules/clusters but also for polystyrene (PS) particles. The particles are closely packed at crystal edges despite that the crystal surface is not illuminated by the laser directly. The molecules/clusters are also gathered and adsorbed to the crystal surface, leading to crystal growth. The trapping dynamics and mechanism are discussed in view of optical potential formed at the crystal surface by light propagation inside the crystal.



SECTION: Surfaces, Interfaces, Porous Materials, and Catalysis

Appl. Phys. B (2013) 112:473–477
DOI 10.1007/s00340-013-5595-y

Appl. Phys. B
DOI 10.1007/s00340-013-5595-y

Applied Physics B
Lasers and Optics

RAPID COMMUNICATION

Single crystal formation of amino acid with high temporal controllability by combining femtosecond and continuous wave laser trapping

Atsushi Miura · Yan-Hua Huang · Hiroshi Masuhara

Abstract We investigated laser trapping crystallization of glycine by using femtosecond (fs) laser as a trapping light source. Impulsively exerted fs laser pulses crystallized glycine more effectively than that induced by continuous wave (CW) laser trapping. Highly efficient crystallization and crystal growth behavior indicates fs laser irradiation increased the concentration not only at the focal spot, but also around the laser focus. Furthermore, we found that irradiation of fs pulses to CW laser-induced locally high supersaturation region enables immediate crystallization. Spatiotemporally controlled triggering of a single crystal formation with sub-second time resolution has achieved by integrating fs and CW laser trapping techniques.

Photochemical & Photobiological Sciences

PAPER

Cite this: *Photochem. Photobiol. Sci.*, 2014, **13**, 254

Laser trapping-induced crystallization of L-phenylalanine through its high-concentration domain formation†

Ken-ichi Yuyama,[a] Chi-Shiun Wu,[a] Teruki Sugiyama*[b] and Hiroshi Masuhara*[a]

Received 9th August 2013,
Accepted 2nd October 2013

DOI: 10.1039/c3pp50276g

www.rsc.org/pps

We present the laser trapping-induced crystallization of L-phenylalanine through high-concentration domain formation in H_2O and D_2O solutions which is achieved by focusing a continuous-wave (CW) near-infrared laser beam at the solution surface. Upon laser irradiation into the H_2O solution, laser trapping of the liquid-like clusters increases the local concentration, accompanying laser heating, and a single plate-like crystal is eventually prepared at the focal spot. On the other hand, in the D_2O solution, a lot of the monohydrate needle-like crystals are observed, not at the focal spot where the concentration is high enough to trigger crystal nucleation, but in the 0.5–1.5 mm range from the focal spot. The dynamics and mechanism of the amazing crystallization behaviour induced by laser trapping are discussed from the viewpoints of the concentration increase due to laser heating depending on solvent, the large high-concentration domain formation by laser trapping of liquid-like clusters, and the orientational disorder of molecules/clusters at the domain edge.

Cryst. Growth Des. 2014, 14, 15−22

CRYSTAL GROWTH & DESIGN

Communication

pubs.acs.org/crystal

Crystal Growth of Lysozyme Controlled by Laser Trapping

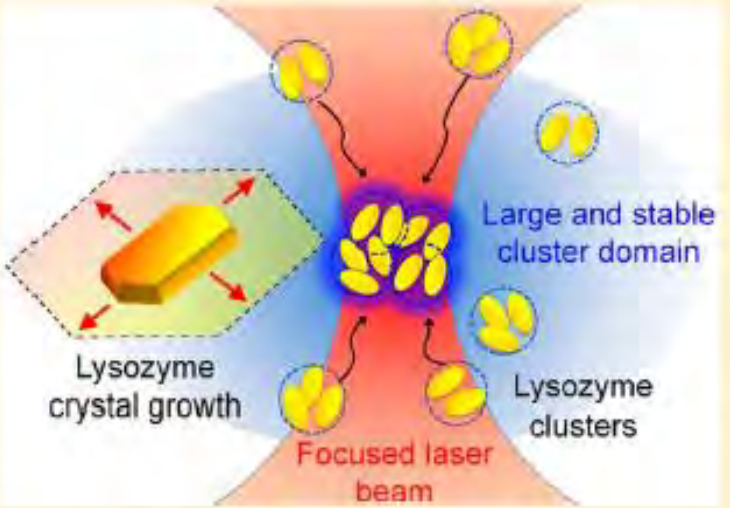
Jing-Ru Tu,[†] Atsushi Miura,[†] Ken-ichi Yuyama,*[,†] Hiroshi Masuhara,*[,†] and Teruki Sugiyama*[,‡]

[†]Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, Hsinchu 30010, Taiwan

[‡]Instrument Technology Research Center, National Applied Research Laboratories, Hsinchu 30076, Taiwan

Supporting Information

ABSTRACT: We demonstrate the growth of a tetragonal crystal of hen egg-white lysozyme (HEWL) in D_2O buffer solution controlled by laser trapping with a focused continuous-wave (CW) near-infrared (NIR) laser beam. The focal spot was located at 10 μm away from the edge of the target crystal that was generated spontaneously, and the crystal growth was observed although the focal spot size was much smaller than the distance. The growth rate of (101) and {110} faces of the tetragonal crystal was examined with various laser powers and polarizations. The rate observed under the irradiation was much different from those in spontaneous growth, namely, the growth rate of the {110} face showed a large decrease or increase depending on the irradiation time. The dynamics and mechanism of this unusual crystal growth behavior is discussed from the viewpoint of a large stable domain formation of the HEWL liquidlike clusters through liquid nucleation and growth and by considering the anisotropy of the cluster domain.



CRYSTAL GROWTH & DESIGN

Article

pubs.acs.org/crystal

Dynamics and Mechanism of Laser Trapping-Induced Crystal Growth of Hen Egg White Lysozyme

Published as part of the Crystal Growth & Design virtual special issue of selected papers presented at the 11th International Workshop on the Crystal Growth of Organic Materials (CGOM11 Nara, Japan), a joint meeting with Asian Crystallization Technology Symposium (ACTS 2014)

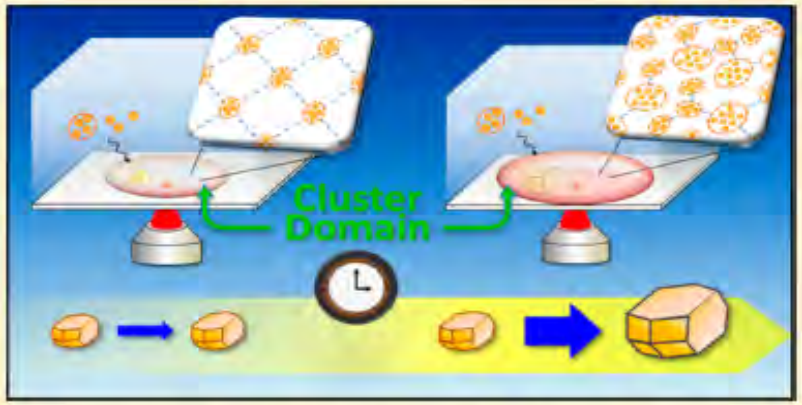
Jing-Ru Tu,[†] Ken-ichi Yuyama,[*,†] Hiroshi Masuhara,[*,†] and Teruki Sugiyama[*,†,‡]

[†]Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, Hsinchu 30010, Taiwan

[‡]Instrument Technology Research Center, National Applied Research Laboratories, Hsinchu 30076, Taiwan

Supporting Information

ABSTRACT: We propose the dynamics and mechanism of laser trapping-induced crystal growth of hen egg-white lysozyme (HEWL). A continuous-wave near-infrared laser beam is used as a trapping light source and focused at a point 10 μm away from a target tetragonal HEWL crystal that is spontaneously generated in solution. Laser trapping of HEWL liquid-like clusters in solution increases local concentration in the focus, where the free motion and orientation of the clusters are strongly restricted, and the clusters show high rigidity and ordering. The cluster association and reorientation at the micrometer-sized focus is evolved to a large highly concentrated domain of the clusters, where the specific target crystal is grown. Initially, the high rigidity and ordering of the clusters strongly suppress the crystal grow rate compared to spontaneous crystal growth. Continuous laser trapping at the focus of the initially formed domain, however, leads to the transition to another domain with different concentration, rigidity, and ordering of the clusters, which surprisingly enhances the crystal growth rate. More interestingly, the clusters in both domains have anisotropic features reflecting the laser polarization direction, which also contributes to the crystal growth.



Cryst. Growth Des. 2015, 15, 4760−4767

PCCP

ROYAL SOCIETY OF CHEMISTRY

COMMUNICATION

Cite this: *Phys. Chem. Chem. Phys.*, 2018, **20**, 6034

Received 13th October 2017,
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DOI: 10.1039/c7cp06990a

Rapid localized crystallization of lysozyme by laser trapping†

Ken-ichi Yuyama, ‡[a] Kai-Di Chang,[a] Jing-Ru Tu,[a] Hiroshi Masuhara [a] and Teruki Sugiyama *[ab]

Applied Physics Express 11, 085502 (2018)
https://doi.org/10.7567/APEX.11.085502

Bubble generation and molecular crystallization at solution surface by intense continuous-wave laser irradiation

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[2]*Research Institute for Electronic Science, Hokkaido University, Sapporo 001-0020, Japan*
[3]*Center for Emergent Functional Matter Science, National Chiao Tung University, Hsinchu 30010, Taiwan*
[4]*Division of Materials Science, Graduate School of Science and Technology, Nara Institute of Science and Technology, Ikoma, Nara 630-0192, Japan*

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We demonstrate bubble generation outside the focus induced by irradiating a focused 1064 nm continuous-wave laser beam into the surface of water and L-phenylalanine H_2O solutions. In the former case of water, bubbles stay at positions distant from the focus during the irradiation, and their size and location are controllable by the laser power. In the latter solution, bubbles move outward toward the surrounding area, and subsequently crystallization takes place at the focus. We discuss these behaviors from the viewpoints of the temperature elevation accompanying the decrease in air solubility as well as the optical trapping of L-phenylalanine clusters giving a single crystal.

CRYSTAL GROWTH & DESIGN

Article

Cite This: Cryst. Growth Des. 2018, 18, 5417−5425

pubs.acs.org/crystal

Pseudopolymorph Control of L-Phenylalanine Achieved by Laser Trapping

Chi-Shiun Wu,[†] Pei-Yun Hsieh,[†] Ken-ichi Yuyama,[†,‡] Hiroshi Masuhara,[†,§] and Teruki Sugiyama*[,†,§,#]

[†]Department of Applied Chemistry and [§]Center for Emergent Functional Matter Science, National Chiao Tung University, Hsinchu 30010, Taiwan

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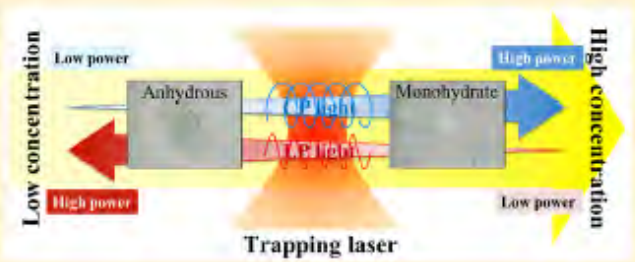
[#]Division of Materials Science, Graduate School of Science and Technology, Nara Institute of Science and Technology, Ikoma 630-0192, Japan

Supporting Information

ABSTRACT: We demonstrate pseudopolymorph control of L-phenylalanine (L-Phe) with laser trapping method by tuning laser power, polarization, and initial solution concentration. When a continuous-wave near-infrared laser beam of 1064 nm was focused at an air/solution interface of L-Phe D_2O solutions with different saturation, L-Phe crystallization was always observed within 30 min-irradiation, even from unsaturated solution. Either whisker- or plate-like crystals were generated depending on given conditions, and identified by Raman spectroscopy to be two L-Phe pseudopolymorphs of monohydrate and anhydrous forms, respectively. The absolute control of L-Phe pseudopolymorphism was achieved by changing the initial solution concentration. In unsaturated solution, laser trapping always produced only one anhydrous crystal at the focus, which can never be produced on spontaneous nucleation at ordinary temperatures and pressures, while in supersaturated solution, a number of monohydrate crystals were densely distributed in an area ranging from 500 μm to 1 mm away from the focus. Moreover, in saturated solution, laser power and polarization contributed to the pseudopolymorphism. As laser power was increased, linearly and circularly polarized laser irradiation increased the formation probability of the anhydrous and monohydrate crystals, respectively. The dynamics and mechanism of laser trapping-induced pseudopolymorphism of L-Phe are discussed in view of the formation of a highly concentrated domain consisting of the liquid-like clusters and the stability of the clusters in the domain under electromagnetic field of trapping laser.



CRYSTAL GROWTH & DESIGN

Article

Crystal Growth and Dissolution Dynamics of L-Phenylalanine Controlled by Solution Surface Laser Trapping

Ken-ichi Yuyama, Ding-Shiang Chiu, Yen-En Liu, Teruki Sugiyama, and Hiroshi Masuhara

Cryst. Growth Des., **Just Accepted Manuscript** • DOI: 10.1021/acs.cgd.8b01233 • Publication Date (Web): 02 Oct 2018

Reviews and Accounts

Pure Appl. Chem., 2011, 83, 869-883.

Laser-trapping assembling dynamics of molecules and proteins at surface and interface*

Hiroshi Masuhara[1,2,‡], Teruki Sugiyama[1], Thitiporn Rungsimanon[1], Ken-ichi Yuyama[1], Atsushi Miura[2], and Jing-Ru Tu[2]

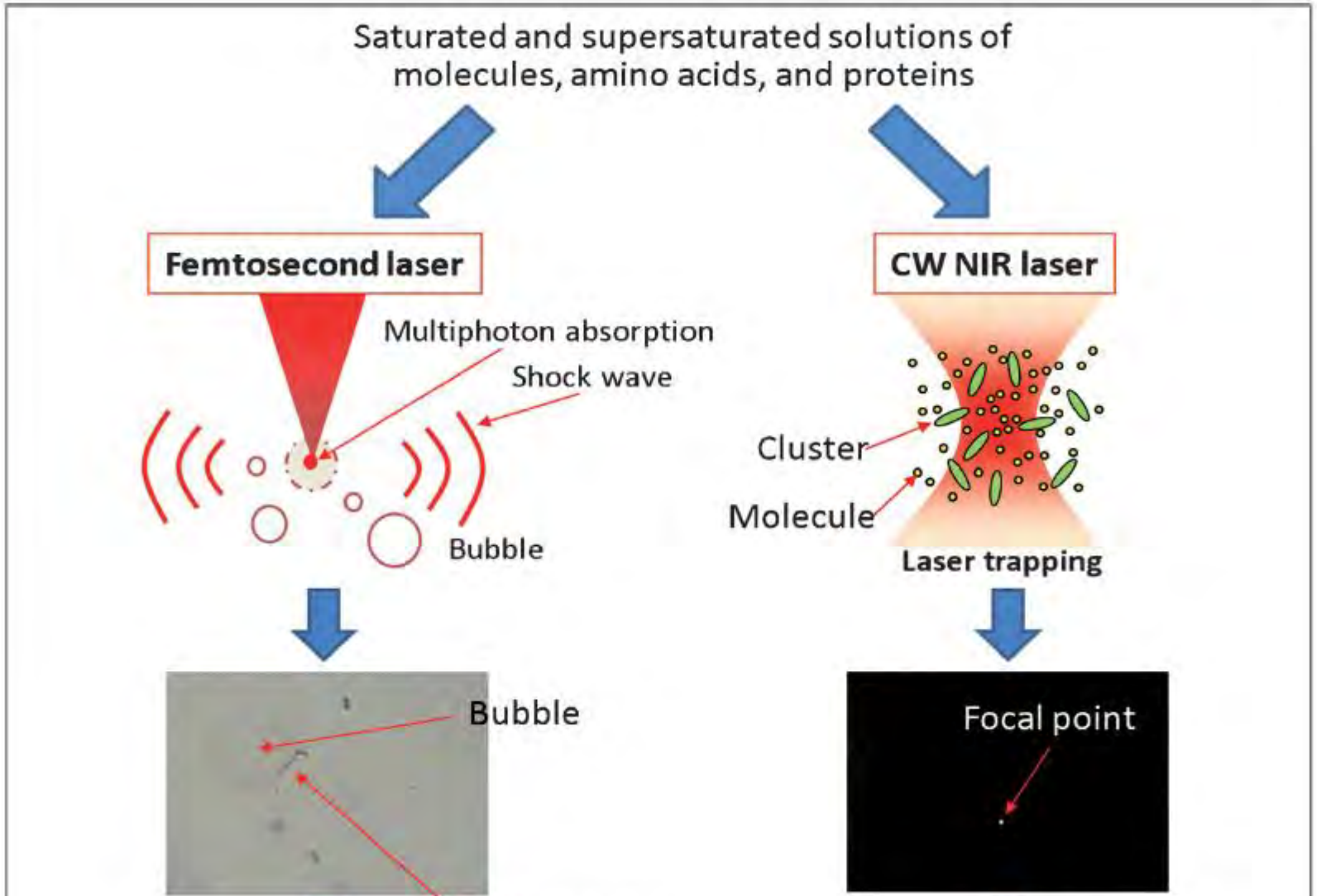
[1]Graduate School of Materials Science, Nara Institute of Science and Technology, Ikoma 630-0192, Japan; [2]Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, Hsinchu 30100, Taiwan

Abstract: Laser trapping of molecules and proteins in solution at room temperature is made possible by irradiating 1064-nm continuous-wave (CW) laser with power around 1 W. Although conventional small molecules are not trapped at the focal point, molecules that can form clusters upon assembling and proteins whose size is close to 10 nm are gathered, giving unique assembly structure. Glycine in H_2O shows crystallization, urea in D_2O gives a millimeter-sized giant droplet, and cobalt oxide-filled ferritin protein confirms assembly followed by precipitation. Solute concentration, solvent, and laser power are important factors for determining trapping and assembling phenomena, and the laser focal position is very critical. These unique behaviors are realized by setting the irradiation at the air/solution surface, inside the solution, and at the glass/solution interface. Laser trapping-induced crystallization, liquid/liquid phase separation, and precipitation are compared with the previous results and considered. After summarizing the results, we describe our future perspective and plans.

Laser-Induced Crystallization and Crystal Growth

Teruki Sugiyama*[a, b] and Hiroshi Masuhara*[c, d]

Chem. Asian J. 2011, 6, 2878 – 2889



ACCOUNTS of chemical research

1946 ■ ACCOUNTS OF CHEMICAL RESEARCH ■ 1946–1954 ■ 2012 ■ Vol. 45, No. 11

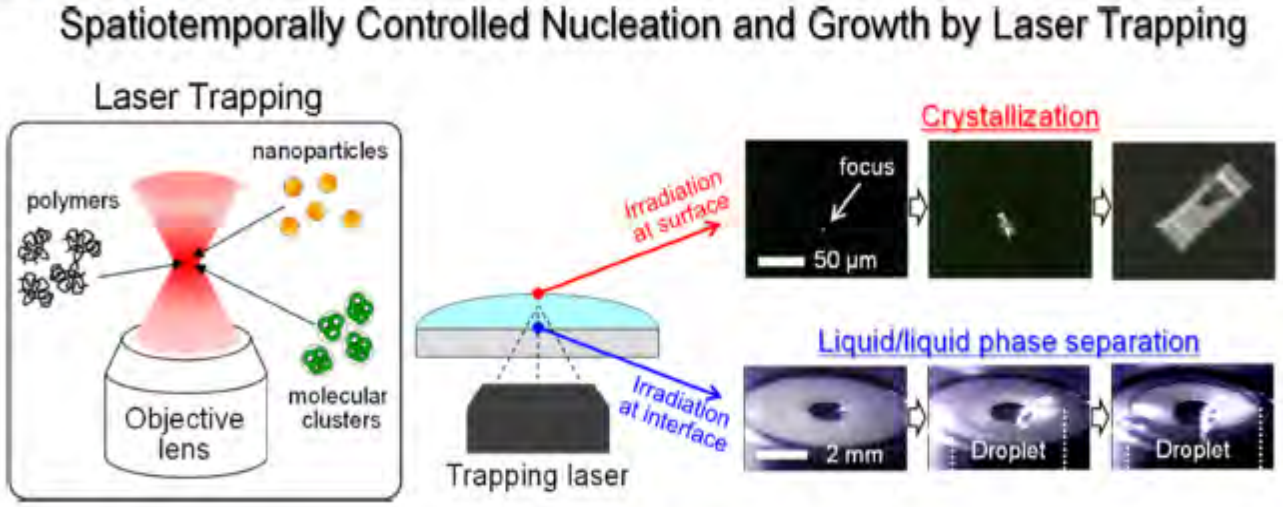
Laser Trapping Chemistry: From Polymer Assembly to Amino Acid Crystallization

TERUKI SUGIYAMA,*,† KEN-ICHI YUYAMA,§ AND HIROSHI MASUHARA*,§

†Instrument Technology Research Center, National Applied Research Laboratories, Hsinchu 30076, Taiwan, and §Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, Hsinchu 30010, Taiwan

RECEIVED ON MAY 29, 2012

CONSPECTUS



Opt Rev (2015) 22:143–148
DOI 10.1007/s10043-015-0029-1

SPECIAL SECTION: INVITED REVIEW PAPER

1st Optical Manipulation Conference (OMC'14), Yokohama, Japan

Optical trapping assembling of clusters and nanoparticles in solution by CW and femtosecond lasers

Hiroshi Masuhara · Teruki Sugiyama · Ken-ichi Yuyama · Anwar Usman

BBA - General Subjects 1862 (2018) 335–357

Contents lists available at ScienceDirect

BBA - General Subjects

journal homepage: www.elsevier.com/locate/bbagen

Review

Novel physical chemistry approaches in biophysical researches with advanced application of lasers: Detection and manipulation☆

Koichi Iwata[a,*], Masahide Terazima[b,*], Hiroshi Masuhara[c,*]

[a] Department of Chemistry, Faculty of Science, Gakushuin University, 1-5-1 Mejiro, Toshima-ku, Tokyo 171-8588, Japan
[b] Department of Chemistry, Graduate School of Science, Kyoto University, Sakyo-ku, Kyoto 606-8502, Japan
[c] Department of Applied Chemistry, National Chiao Tung University, 1001 Ta Hsueh Rd., Hsinchu 30010, Taiwan

Exploring New Trapping Phenomena

Langmuir 2017, 33, 755−763

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Article

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Photocontrolled Supramolecular Assembling of Azobenzene-Based Biscalix[4]arenes upon Starting and Stopping Laser Trapping

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Supporting Information

ABSTRACT: Laser trapping in chemistry covers various studies ranging from single molecules, nanoparticles, and quantum dots to crystallization and liquid−liquid phase separation of amino acids. In this work, a supramolecular assembly of azobenzene-based biscalix[4]arene is generated in ethyl acetate using laser trapping; its nucleation and growth are elucidated. No trapping behavior was observed when a 1064 nm laser beam was focused inside of the solution; however, interesting assembling phenomena were induced when it was shined at the air/solution interface. A single disk having two layers was first prepared at the focal point of ∼1 μm and then expanded to the size of a few tens of micrometers, although no optical force was exerted outside of the focal volume. Upon switching the trapping laser off, needles were generated at the outer layer of the assembly, giving a stable sea urchin-like morphology to the generated assembly. At a 30−50% dilution of the initial solution in ethyl acetate, a mushroom-like morphology was also observed. Laser trapping-induced assembly of azobenzene-based biscalix[4]arene was quite different from the sharp-ellipsoidal aggregates obtained by the spontaneous evaporation of the solution. These trapping phenomena were specifically observed for biscalix[4]arene in the *trans* conformation of azo-benzene moiety but not for the *cis*-form, suggesting that the laser trapping of this azobenzene-based biscalix[4]arene is photocontrollable. Dynamics and mechanism of the supramolecular assembling are considered, referring to laser trapping-induced nucleation and liquid−liquid phase separation of amino acids.

Angew. Chem. Int. Ed. 2017, 56, 6739 –6743

Amyloid Fibrils Hot Paper

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German Edition: DOI: 10.1002/ange.201702352

A Single Spherical Assembly of Protein Amyloid Fibrils Formed by Laser Trapping

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Abstract: *Protein amyloids have received much attention owing to their correlation with serious diseases and to their promising mechanical and optical properties as future materials. Amyloid formation has been conducted by tuning temperature and chemical conditions, so that its nucleation and the following growth are analyzed as ensemble dynamics. A single spherical assembly of amyloid fibrils of cytochrome c domain-swapped dimer was successfully generated upon laser trapping. The amyloid fibrillar structure was confirmed by fluorescence characterization and electron microscopy. The prepared spheres were further manipulated individually in solution to fabricate a three-dimensional microstructure and a line pattern.*

magnetic field,[4] and pulsed laser irradiation.[5,6] Therefore, all the processes of nucleation and growth of amyloid and crystal in solution proceed randomly in parallel, whose dynamic evolutions are monitored and analyzed as an ensemble of amyloid fibrils or crystals. It is considered very promising to propose a new experimental approach for preparing a single spherical assembly of protein amyloid, analyzing its dynamics, and fabricating micro-structures from single assemblies. It will enable us to perform amyloid studies by watching always when and where individual assemblies of amyloid fibrils are prepared, monitored, and utilized.

Herein we report a laser trapping study on oxidized

Japanese Journal of Applied Physics
Vol. 45, No. 16, 2006, pp. L453–L456

Enhancement of Biased Diffusion of Dye-Doped Nanoparticles by Simultaneous Irradiation with Resonance and Nonresonance Laser Beams

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We propose and demonstrate the enhancement of the biased diffusion of dye-doped nanoparticles using resonance and nonresonance laser beams. The Brownian motion of nanoparticles in a laser focus is investigated by fluorescence correlation spectroscopy (FCS) and the time variation in fluorescence intensity. From the analysis of autocorrelation functions, it is demonstrated that the difference between the transit times of nanoparticles in the focal spot with and without resonance laser irradiation increases ~7-fold by the simultaneous irradiation of a near-infrared laser. This method is applicable to the selective optical manipulation of dye-stained nanomaterials and biomolecules in solution. [DOI: 10.1143/JJAP.45.L453]

KEYWORDS: optical trapping, nanoparticle, fluorescence correlation spectroscopy

Laser Phys. Lett. **11** (2014) 076001 (8pp)
doi:10.1088/1612-2011/11/7/076001

Enhanced optical confinement of dye-doped dielectric nanoparticles using a picosecond-pulsed near-infrared laser

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Abstract

We demonstrate a novel strategy to increase the capability of confining numerous dye-doped polymeric nanobeads (diameter 100 nm) with laser trapping. Unlike most classical works of optical trapping that address mainly the stiffness of the optical trap, our work concerns an increase in the number of particles confined near the laser focus. We developed an imaging system of light scattering in which a condenser lamp was employed to illuminate the focal plane of the objective lens, and the scattering of the incoherent light was specifically measured to determine the number of confined nanobeads. In contrast to preceding work that used mainly continuous-wave or femtosecond-pulsed lasers, we employed a picosecond-pulsed laser with the half-wavelength of the laser particularly falling within the absorption band of the dopant. Our results show that the number of doped nanobeads held by the laser

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PAPER

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Enhanced optical confinement of dielectric nanoparticles by two-photon resonance transition†

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Despite a tremendous success in the optical manipulation of microscopic particles, it remains a challenge to manipulate nanoparticles especially as the polarizability of the particles is small. With a picosecond-pulsed near-infrared laser, we demonstrated recently that the confinement of dye-doped polystyrene nanobeads is significantly enhanced relative to bare nanobeads of the same dimension. We attributed the enhancement to an additional term of the refractive index, which results from two-photon resonance between the dopant and the optical field. The optical confinement is profoundly enhanced as the half-wavelength of the laser falls either on the red side, or slightly away from the blue side, of the absorption band of the dopant. In contrast, the ability to confine the nanobeads is significantly diminished as the half-wavelength of the laser locates either at the peak, or on the blue side, of the absorption band. We suggest that the dispersively shaped polarizability of the dopant near the resonance is responsible to the distinctive spectral dependence of the optical confinement of nanobeads. This work advances our understanding of the underlying mechanism of the enhanced optical confinement of doped nanoparticles with a near-infrared pulsed laser, and might facilitate future research that benefits from effective sorting of selected nanoparticles beyond the limitations of previous approaches.

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Resonance optical trapping of individual dye-doped polystyrene particles with blue- and red-detuned lasers

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Abstract: We demonstrate resonance optical trapping of individual dye-doped polystyrene particles with blue- and red-detuned lasers whose energy are higher and lower compared to electronic transition of the dye molecules, respectively. Through the measurement on how long individual particles are trapped at the focus, we here show that immobilization time of dye-doped particles becomes longer than that of bare ones. We directly confirm that the immobilization time of dye-doped particles trapped by the blue-detuned laser becomes longer than that by the red-detuned one. These findings are well interpreted by our previous theoretical proposal based on nonlinear optical response under intense laser field. It is discussed that the present result is an important step toward efficient and selective manipulation of molecules, quantum dots, nanoparticles, and various nanomaterials based on their quantum mechanical properties.

PHYSICAL REVIEW E 70, 061410 (2004)

Optical assembling dynamics of individual polymer nanospheres investigated by single-particle fluorescence detection

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(Received 6 July 2004; published 22 December 2004)

When a laser beam is focused into colloidal nanoparticle suspensions, a number of nanoparticles can be confined in the focal spot due to an optical gradient force. To reveal the assembling dynamics of polymer nanoparticles, the assembling process was investigated by analyzing the time evolution of the fluorescence intensity of the nanoparticles. In a dilute suspension of 100-nm-sized particles, a stepwise increase of the fluorescence intensity corresponding to a trapped single nanoparticle was observed. Statistical analysis revealed that the initial assembling rate of nanoparticles was proportional to the laser power and concentration of particle suspensions as expected from the diffusion equation. In 40-nm-sized particle suspensions, blinking profiles of fluorescence intensity were obtained, in which 2–3 particles were simultaneously trapped and then escaped from the focal point. It is considered from statistical analyses and two-dimensional Monte Carlo simulations that this assembling phenomenon is attributable to cluster formation assisted by optical trapping.

PHYSICAL REVIEW E 72, 021408 (2005)

Cluster formation of nanoparticles in an optical trap studied by fluorescence correlation spectroscopy

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(Received 20 April 2005; revised manuscript received 14 June 2005; published 29 August 2005)

We report *in situ* observation of cluster growth of nanoparticles confined in an optical trapping potential by means of fluorescence correlation spectroscopy. When an optical trapping force caused by a highly focused laser beam acts on nanoparticle suspensions, the number of nanoparticles increases and an assembly can be formed at the focal spot. The decay times of fluorescence autocorrelation curves were investigated as a function of the irradiation time of the laser beam and the laser power. In the initial stage of the optical assembling, the decay time increases with the irradiation time of the laser beam. On the other hand, in the later stage, a decrease of the decay time was observed. This behavior is explained successfully by using two models of Brownian motion under weak and strong optical trapping. It was revealed that trapping and clustering of nanoparticles proceed simultaneously and clusters confined in the focal spot make larger aggregates spontaneously.

J. Phys. Chem. C 2016, 120, 15578−15585

Reflection Microspectroscopic Study of Laser Trapping Assembling of Polystyrene Nanoparticles at Air/Solution Interface

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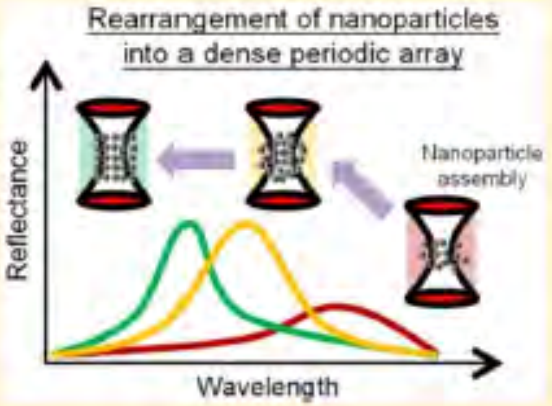
Supporting Information

ABSTRACT: We present the formation of a single nanoparticle assembly with periodic array structure induced by laser trapping of 200 nm polystyrene nanoparticles at air/solution interface of the colloidal heavy water solution. Their trapping and assembling behavior is observed by monitoring transmission and backscattering images and measuring reflection spectra under a microscope. Upon the laser irradiation into the solution surface layer, nanoparticles are gathered at and around the focal spot, and eventually a nanoparticle assembly with the size much larger than the focal volume is formed. The assembly gives structural color in visible range under halogen lamp illumination, indicating that constituent nanoparticles are periodically arrayed. Reflection spectra of the assembly show a reflection band, and its peak position is gradually shifted to short wavelength and the bandwidth becomes narrow with time, depending on the distance from the focal spot. After the laser is switched off, red-shift is observed in the reflection band. These results indicate that nanoparticles are rearranged into a densely packed periodic array during laser irradiation and diffused out to the surrounding solution after turning off the laser. These dynamics are discussed from the viewpoints of the attractive optical trapping force and the electrostatic repulsive force among nanoparticles.



Langmuir 2016, 32, 12488−12496

Optically Evolved Assembly Formation in Laser Trapping of Polystyrene Nanoparticles at Solution Surface

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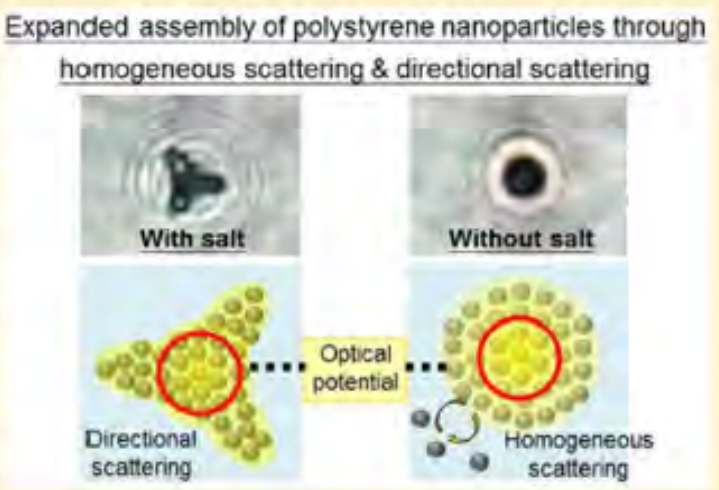
Supporting Information

ABSTRACT: Assembling dynamics of polystyrene nanoparticles by optical trapping is studied with utilizing transmission/reflection microscopy and reflection microspectroscopy. A single nanoparticle assembly with periodic structure is formed upon the focused laser irradiation at solution surface layer and continuously grows up to a steady state within few minutes. By controlling nanoparticle and salt concentrations in the colloidal solution, the assembling behavior is obviously changed. In the high concentration of nanoparticles, the assembly formation exhibits fast growth, gives large saturation size, and leads to dense packing structure. In the presence of salt, one assembly with the elongated aggregates was generated from the focal spot and 1064 nm trapping light was scattered outwardly with directions, while a small circular assembly and symmetrical expansion of the 1064 nm light were found without salt. The present nanoparticle assembling in optical trapping is driven through multiple scattering in gathered nanoparticles and directional scattering along the elongated aggregates derived from optical association of nanoparticles, which dynamic phenomenon is called optically evolved assembling. Repetitive trapping and release processes of nanoparticles between the assembly and the surrounding solution always proceed, and the steady state at the circular assembly formed by laser trapping is determined under optical and chemical equilibrium.



Nano Lett. 2016, 16, 3058−3062

Optical Trapping-Formed Colloidal Assembly with Horns Extended to the Outside of a Focus through Light Propagation

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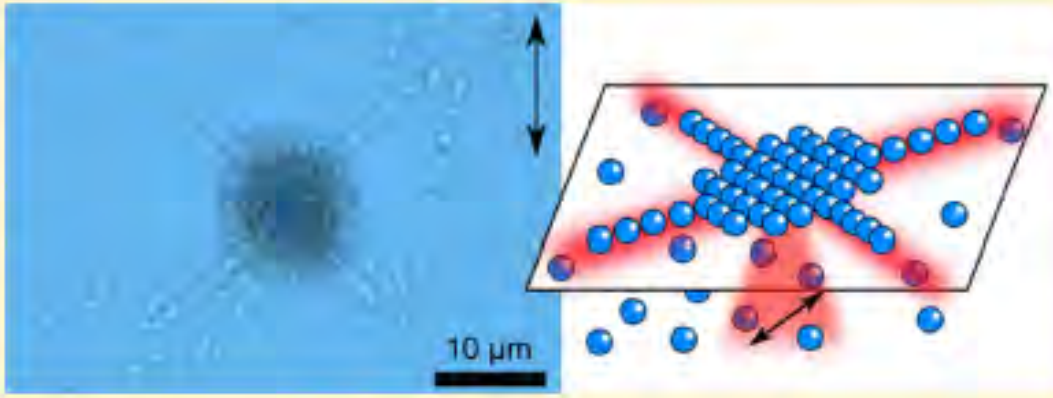
Supporting Information

ABSTRACT: We report optical trapping and assembling of colloidal particles at a glass/solution interface with a tightly focused laser beam of high intensity. It is generally believed that the particles are gathered only in an irradiated area where optical force is exerted on the particles by laser beam. Here we demonstrate that, the propagation of trapping laser from the focus to the outside of the formed assembly leads to expansion of the assembly much larger than the irradiated area with sticking out rows of linearly aligned particles like horns. The shape of the assembly, its structure, and the number of horns can be controlled by laser polarization. Optical trapping study utilizing the light propagation will open a new avenue for assembling and crystallizing quantum dots, metal nanoparticles, molecular clusters, proteins, and DNA.



KEYWORDS: *Optical trapping, colloidal assembly, light propagation, optical binding, glass/solution interface*

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A Single Large Assembly with Dynamically Fluctuating Swarms of Gold Nanoparticles Formed by Trapping Laser

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Supporting Information

ABSTRACT: Laser trapping has been utilized as tweezers to three-dimensionally trap nanoscale objects and has provided significant impacts in nanoscience and nanotechnology. The objects are immobilized at the position where the tightly focused laser beam is irradiated. Here, we report the swarming of gold nanoparticles in which component nanoparticles dynamically interact with each other, keeping their long interparticle distance around the trapping laser focus at a glass/solution interface. A pair of swarms are directionally extended outside the focal spot perpendicular to the linear polarization like a radiation pattern of dipole scattering, while a doughnut-shaped swarm is prepared by circularly polarized trapping laser. The light field is expanded as scattered light through trapped nanoparticles; this modified light field further traps the nanoparticles, and scattering and trapping cooperatively develop. Due to these nonlinear dynamic processes, the dynamically fluctuating swarms are evolved up to tens of micrometers. This finding will open the way to create various swarms of nanoscale objects that interact and bind through the scattered light depending on the properties of the laser beam and the nanomaterials.

KEYWORDS: *Laser trapping, gold nanoparticles, interface, optical binding, light scattering, swarming*

Our Study
on
Femtosecond Laser Trapping Dynamics of Nanoparticles

Journal of Photochemistry and Photobiology A: Chemistry 234 (2012) 83–90

Optical trapping and polarization-controlled scattering of dielectric spherical nanoparticles by femtosecond laser pulses

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ABSTRACT

We present optical trapping behavior of 50-nm-sized polystyrene beads, suspended in water medium, by femtosecond pulsed laser beam. In addition to a higher number of nanoparticles trapped at the focal spot by the ultrashort laser pulses compared with that by continuous-wave laser, the nanoparticles are scattered out of the focal spot by the laser pulses to the surrounding area. The scattered particles form a partially opened folding fan-shaped bright locus in two opposite directions, in an alternating manner, perpendicular to the laser polarization. To understand those phenomena, we analyzed radiation (gradient and scattering) force of femtosecond laser pulses and their temporal force exerted on the dielectric spherical nanoparticles by taking into account the impulsive peak power and the axial component of electric light field produced by high numerical aperture of objective lens. We show that the axial electric field is responsible for lateral components of the scattering and temporal forces, and hence, controls the scattering directions of the Rayleigh particles. These findings provide important information about the dynamic optical trapping of the Rayleigh particles by highly focused ultrashort laser pulses.

J. Phys. Chem. C 2013, 117, 19182−19188

Femtosecond Pulse-Width Dependent Trapping and Directional Ejection Dynamics of Dielectric Nanoparticles

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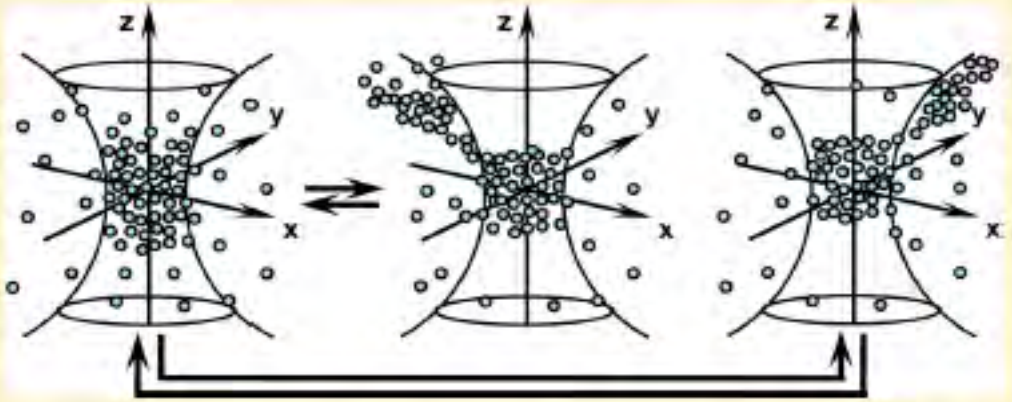
Supporting Information

ABSTRACT: We demonstrate that laser pulse duration, which determines its impulsive peak power, is an effective parameter to control the number of optically trapped dielectric nanoparticles, their ejections along the directions perpendicular to polarization vector, and their migration distances from the trapping site. This ability to controllably confine and eject the nanoparticle is explained by pulse width-dependent optical forces exerted on nanoparticles in the trapping site and ratio between the repulsive and attractive forces. We also show that the directional ejections occur only when the number of nanoparticles confined in the trapping site exceeds a definite threshold. We interpret our data by considering the formation of transient assembly of the optically confined nanoparticles, partial ejection of the assembly, and subsequent filling of the trapping site. The understanding of optical trapping and directional ejections by ultrashort laser pulses paves the way to optically controlled manipulation and sorting of nanoparticles.



Optical trapping of nanoparticles by ultrashort laser pulses

Science Progress (2013), 96(1), 1–18

ANWAR USMAN, WEI-YI CHIANG and HIROSHI MASUHARA

Dr Anwar Usman gained his PhD from Tohoku University (Japan), and extended his researches in several places including Universiti Sains Malaysia, Max-Born-Insitut für Kurzzeitspektroskopie im Forschungsverbund Berlin, Osaka University, and École Normale Supérieure de Chimie Paris. He is now a research fellow working with the Masuhara group in the Laser Bio/Nano Science Laboratory of National Chiao Tung University, which is supported by a project on laser trapping accompanying photochemical and photophysical processes financed by National Science Council of Taiwan. E-mail: usman@faculty.nctu.edu.tw

Wei-Yi Chiang is a PhD student in the Department of Applied Chemistry, National Chiao Tung University. His research is supported by a project operated in the Masuhara group. He is carrying out research on laser trapping with femtosecond laser pulses on target particles ranging from dielectric nanoparticles, nanocrystals, and quantum dots.
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Dr Hiroshi Masuhara graduated from Tohoku University (1966) and obtained his PhD degree from Osaka University (1971). After his retirement from Osaka University, he joined the Department of Applied Chemistry and Institute of Molecular Science of the National Chiao Tung University in Taiwan as a Chair Professor. He is foreign fellow of the National Academy of Sciences India and of the Royal Flemish Academy of Belgium and received the Asian Photochemistry Association Award

Phys. Chem. B 2014, 118, 14010−14016

Efficient Optical Trapping of CdTe Quantum Dots by Femtosecond Laser Pulses

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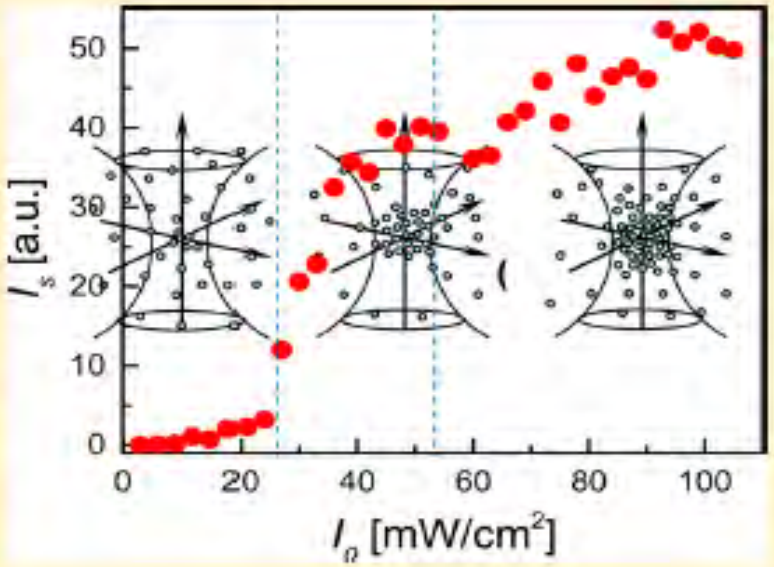
ABSTRACT: The development in optical trapping and manipulation has been showing rapid progress, most of it is in the small particle sizes in nanometer scales, substituting the conventional continuous-wave lasers with high-repetition-rate ultrashort laser pulse train and nonlinear optical effects. Here, we evaluate two-photon absorption in optical trapping of 2.7 nm-sized CdTe quantum dots (QDs) with high-repetition-rate femtosecond pulse train by probing laser intensity dependence of both Rayleigh scattering image and the two-photon-induced luminescence spectrum of the optically trapped QDs. The Rayleigh scattering imaging indicates that the two-photon absorption (TPA) process enhances trapping ability of the QDs. Similarly, a nonlinear increase of the two-photon-induced luminescence with the incident laser intensity fairly indicates the existence of the TPA process.



J. Phys. Chem. C 2016, 120, 5251−5256

Picosecond Motional Relaxation of Nanoparticles in Femtosecond Laser Trapping

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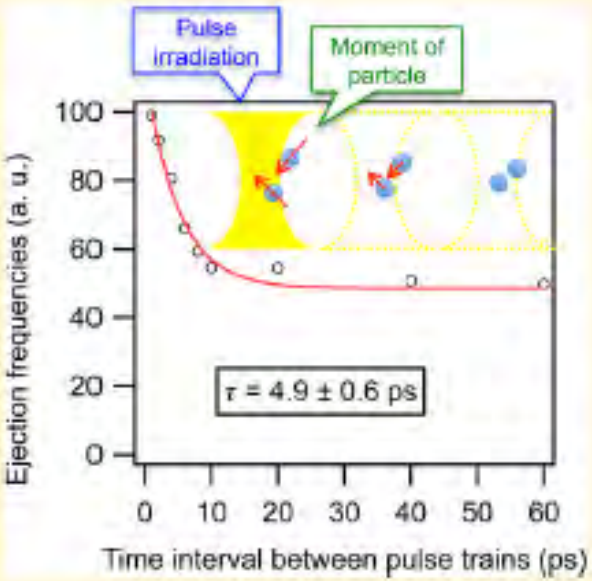
ABSTRACT: Repetitive drag and release dynamics by impulsive force is characteristic of optical trapping by femtosecond laser pulses. We studied the dynamics utilizing double pulse train and found that trapped polystyrene particles are ejected repetitively from the focal spot and its frequencies become less for longer interval of the pulse trains. The ejection changes drastically in a few-ps interval region, although particles cannot move appreciable distance in such a short time. It means that displacement of particles by a conventional diffusive motion is not dominant and another fast process has an important role in femtosecond pulse trapping. We also revealed that the silica nanoparticles shows a decay at few-ps, indicating that the picosecond decay is not due to a material property but considered to be a general dynamics. We propose that a picosecond relaxation process of inertia force of particles is important for understanding laser trapping dynamics by femtosecond laser pulses.



J. Phys. Chem. C 2016, 120, 2392−2399

Optical Trapping Dynamics of a Single Polystyrene Sphere: Continuous Wave versus Femtosecond Lasers

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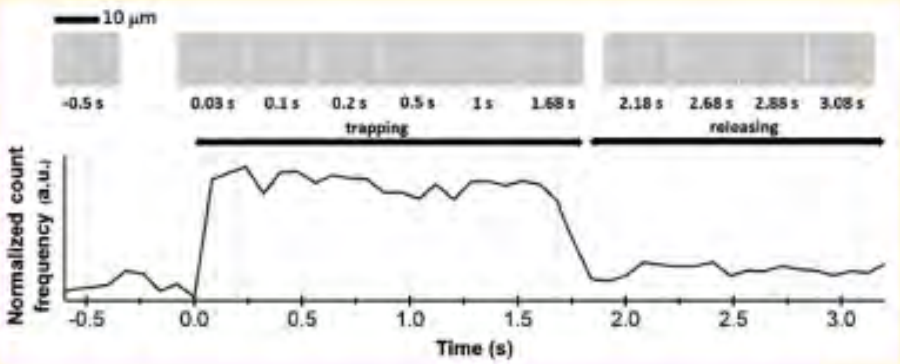
ABSTRACT: Understanding of optical trapping dynamics of a single particle in the trapping site is important to develop its optical manipulation for molecular assembly and chemical application. For micrometer-sized Mie particles, similar trapping efficiency of the conventional continuous wave (cw) laser or high-repetition-rate femtosecond (fs) laser pulse train has been established [Dholakia et al., *Opt. Express* **2010**, *18*, 7554−7568], in contrast to higher efficiency of the laser pulses to trap dielectric Rayleigh particles. To further explore and clarify the switching phenomena of optical trapping efficiency with cw laser and fs laser pulse and to elucidate its nature, we study the immobilization dynamics of a single polystyrene sphere with 500 nm in diameter (which is comparable to focal beam size) in shallow potential well. By observing trapping events and immobilization time of the particle with a size in Lorenz−Mie regime, distinct from well-known Rayleigh particle and ray optics approximations, we found that immobilization time is only linearly related to the incident laser power ≤40 mW, and at higher laser powers cw laser is more efficient than fs laser pulses to immobilize the particle. This finding means that the dynamics of the particle in this size region is still affected by the strong transient force fields induced by high-repetition-rate ultrashort pulse train as usually observed for Rayleigh particles. This may provide an understanding that the dynamics of the target particle in the trapping site is size- and laser mode-dependent.



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THE JOURNAL OF PHYSICAL CHEMISTRY C

Article

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Femtosecond Laser Trapping Dynamics of Nanoparticles: A Single Transient Assembly Formation Leading to Their Directional Ejection

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Supporting Information

ABSTRACT: We investigated femtosecond laser trapping dynamics of silica nanoparticles with different hydrophobic surface properties. We demonstrated that the hydrophobic surface on the silica nanoparticles facilitates mutual association of the nanoparticles in the optical trapping site. Such association of optically trapped nanoparticles is a prerequisite to induce their directional ejection away from the trapping site. The directional ejection of the optically trapped nanoparticles is most probably due to asymmetric three-dimensional ejecting forces generated by the electromagnetic interaction between transient assembly in the focal spot and the incident pulses. These findings provide important insights into the directional ejection of nanoparticles from the trapping site in the femtosecond laser trapping, and this physicochemical phenomenon is controlled by both the trapping laser and material properties.

Our Laser Trapping Study in NCTU for 2008 April - Present

Nanoparticles

Polystyrene
Silica
Gold

Trapping
Manipulation
Assembling

Molecules

Polymer
Supramolecule
Dye
Micelle

Assembling
Growth
Nucleation
Droplet formation
Crystallization
Amyloid fibril formation

Methodology

Spectroscopy
Lasing
Resonance Effect
Femtosecond

Patterning
Deposition

Ablation