Emergence of Exotic Spatio-Temporal Structure under Photon Flux

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Abstract. The operation of laser focusing on an object implies the creation of dielectric dielectric field under a thermodynamically open condition. Thus, we can expect the appearance of the effect of thermal irreversibility, such as breakdown of detailed balance, occurrence of circular state-flux in the phase-space, and limit-cycle oscillation. In the present article, we describe our recent experimental results on various kinds of exotic time-dependent phenomena induced by the continuous irradiation of laser. 1) Generation/annihilation of droplets from binary homogeneous liquid induced by laser: It will be shown that focused laser induces micro-phase separation on an oil/water isotropic solution. By choosing the proper experimental conditions, rhythmic change of generation, growth, and disappearance of a droplet at the focus is generated. This rhythmic phenomenon is a kind of limit-cycle oscillation. 2) Positive/negative photophoresis on a droplet: We show that a droplet is driven by a laser beam, either toward and backward along the direction of photon flux, through the change of the position of irradiation. Such photophoretic motion is induced by interfacial instability owe to the laser irradiation. 3) Rhythmic growth and bursting of a cluster with micro-beads: It is shown that negatively charged micro-beads are collected toward the focus of IR laser, i.e., optical tweezers. When the focusing angle is decreased from usual conditions, rhythmic change of the formation-growth-bursting of the beads cluster is generated.

1. Introduction
Nature creates rich variety of exotic phenomena under far-from-equilibrium conditions, i.e., evolving universe, living organisms, weather, etc. Photo-irradiation exhibits the physical meaning to afford non-equilibribicity with a well-controlled manner. We can think of the fact that lives on the earth maintain their activity under the flux of solar energy. Thus, it may of scientific value to explore the currently unexplored scenario of spatio-temporal structural formation on a thermodynamically open system created under photon-flux. In the present paper, we would like to describe some of our recent experimental examples on the emergence of time-reversal symmetry-breaking on soft-materials caused by laser illumination.

For the actual experimental system, we adapt the experimental condition essentially the same as in usual laser tweezers [1,2]. It has been well established that focused laser generates a potential field on an object. Depending on the size of the object, theoretical interpretation changes either Mie scattering or Rayleigh scattering. In both of the theoretical frameworks, static filed, attractive or repulsive, is
exerted around the focus of the laser. Here, it is of importance to notice that laser is directional flux photons, suggesting that the effect of thermodynamically open condition may induce various kinds of nonequilibrium phenomena. We would like to demonstrate that focused laser serves as a useful experimental tool to create thermodynamically open condition on a local finite-space under a desired condition.

2. Rhythmicity on the Generation/Annihilation of a Droplet

We used an isotropic solution of D$_2$O mixed with triethylamine or 3-methylpyridine, exhibiting a lower critical solution temperature (LCST) type phase-separation around room temperature [3,4,5]. The solutions were situated in a glass chamber and observed with an inverted phase-contrast microscope equipped with an oil-immersed objective lens (100×, N.A.: 1.3). The temperature of the solutions was controlled with the precision of ±0.1K through the use of a temperature control unit. A CW Nd:YAG laser (Spectra Physics, 1064 nm) was introduced into the microscope. Under such a condition, heating by the laser has only a minute effect on the induction of phase-separation.

Figure 1 shows a spatio-temporal image of the emergence/nucleation and successive growth of an oil droplet at the focus of the laser ($\lambda=1064$ nm) in homogeneous medium rich in water at a laser power of 1.5 W, where the actual shapes of the droplets are given at the bottom [3]. Just after the start of irradiation at a certain place in the homogeneous solution, a small droplet appeared, and after ca.80 s, the droplet reached a constant diameter. We found that such stable trapping is observed above 1.4 W. We performed a similar experiment in which H$_2$O was replaced by D$_2$O. It is known that the absorption of the length ($\lambda=1064$ nm) in D$_2$O is less than 1/10 of that in H$_2$O. We have confirmed that there is no essential difference in the experimental trends between H$_2$O and D$_2$O, suggesting that the heating effect of the laser has only minute effect on the induction of microscopic phase separation.

Figure 2 exemplifies the time-successive images near the focus, indicating the occurrence of rhythmicity in the $\mu$m-sized phase-separating pattern in a solution of D$_2$O mixed with 3-methylpyridine (3MP) at a laser power of 67mW [5]. Soon after the homogeneous mixture is irradiated by a laser beam, a 3MP-rich droplet appears at the focal point, and grows larger with time. The droplet spontaneously escapes from the focal point after growing to a certain size, i.e., radius of 13 $\mu$m, and then dissolves into the homogeneous mixture. A new 3MP-rich droplet then emerges at the focal point. This emergence-growth-escape pattern is repeated periodically, like a kind of limit-cycle oscillation. Figure 2(b) shows a spatio-temporal image of this rhythmic phenomenon and the temporal change in the radius. We found that such a rhythmic phenomenon can be observed when the power is above 63 mW. As the laser power increases, the frequency of the emergence growth-escape pattern
also increases. The radius of the droplet just before it is released from the focal point is almost independent of the laser power. When $P < 63 \text{ mW}$, only a homogeneous mixture is observed without the occurrence of phase-separation.

![Figure 2. Rhythmicity of appearance-growth-annihilation of a droplet at the focus under CW laser [5]. (a) Microscopic image together with a schematic illustration the rhythmic change in the phase-separation pattern observed in an isotropic solution of of D$_2$O mixed with 3-methylpyridine (3MP). (b) Spatio-temporal image of the experiment shown in (a).](image)

### 3. Positive/Negative Photophoresis
As for the photophoretic motion, it has been found that some types of objects floating in the gas phase move toward the source of the laser beam, which has been described in terms of radiometric force [6]. Here, we show the successful control of the forward/backward directed motion an oil droplet floating on an aqueous phase [7]. Interesting to say, the direction of the droplet motion can be switched between forward and backward depending on the position of the laser irradiation on the same object. Such controlled motion can be explained in terms of photomechanical energy conversion into mechanical motion through the Marangoni instability [8], i.e., spontaneous surface motion caused by the interfacial tension under temperature gradient. Under local heating on an oil droplet by a narrow laser beam, this local heating induces convection inside the droplet, and the convective motion, thus generated, produces translational directed motion of the droplet.

### 4. Rhythmic growth and bursting of a cluster with micro-bead
In general, the force generated with a focused beam is represented as the summation of gradient and scattering terms [9],

$$\vec{F} = \alpha \nabla \left( \hat{E}^2 \right) + \beta \hat{E} \times \hat{H},$$

where $\hat{E}$ and $\hat{H}$ are the electromagnetic field of the focused light, and $\alpha$ and $\beta$ are constants, as a function of the refractive index of the aqueous medium and the trapped object. The first term describes an attractive potential around the focus, where the attractive force decreases with decrease of the convergence angle of the optical cone. In conventional laser-trap experiments, the convergence angle is maximized in order to make the trapping force larger. In contrast, as in the last term in the above equation, the scattering force exerted on an object increases with decreases in the convergence angle. It is thus expected that instability in optical trapping would be induced by decreases in the convergence angle. Bearing this effect in mind, we have carried out a laser-trapping experiment with submicrometer-sized plastic beads exhibiting a negative charge. Without laser irradiation, the beads disperse homogeneously in aqueous solution due to their negative charge. A converged laser beam
under a standard angle 120° for the optical cone induces stationary clustering of the beads. By changing the convergence angle from 120° to 80°, a rhythmic change between the growth and the bursting of the cluster is generated [10]. Together with theoretical consideration, it is concluded that the observed oscillatory phenomenon is caused by the competition between the trapping and scattering forces exerted by the focused laser, for a system of negatively charged beads that repel each other.

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