Anisotropic dynamics of nanoparticles in clusters at a solid-liquid interface by laser trapping

Itsuo Hanasaki¹, Chie Hosokawa²,³

¹Institute of Engineering, Tokyo University of Agriculture and Technology, Naka-cho 2-24-16, Koganei, Tokyo 184-8588, Japan
²Biomedical Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), 1-8-31 Midorigaoka, Ikeda, Osaka 563-8577, Japan,
³Advanced Photonics and Biosensing Open Innovation Laboratory, AIST, Yamadaoka 2-1, Suita, Osaka 565-0871, Japan
E-mail: hanasaki@cc.tuat.ac.jp

Abstract. It is well-recognized that the Brownian motion of particles in fluids is random. Nevertheless, there can be characteristics depending on the specific physical conditions. We analyze the system of nanoparticle clusters formed by the laser trapping force field at the solid-liquid interface, based on the microscopy movie data. Since the laser trapping force field is basically a function of radial distance from the focal point in the two dimension at the liquid-solid interface, we examine the difference of displacement distributions in the radial and circumferential directions. The results show that the basic characteristics in this system depends on the laser power, and there is an anisotropy in the stochastic motion of the nanoparticles.

1. Introduction

When laser-trapping force field is applied in colloidal dispersion in the vicinity of the solid wall, a cluster of trapped nanoparticles can be obtained. In particular, depending on the details of the laser power etc., ring structures of nanoparticles are formed. The Brownian motion of particles in fluid can exhibit characteristics that originate from this specific configuration. This might affect the initiation events of crystallization by laser-induced perturbation [1]. There are rooms for the analysis beyond the overall diffusion coefficient, depending on the specific physical conditions on either particles themselves [2, 3] or the surrounding environments [4] and available amount as well as the precision of the data. We analyze the movie data obtained from the optical microscopy and camera system. We track the particles in the microscopy movie and evaluate the displacement distribution. There can be effective difference in the nature of Brownian motion in the radial direction and its perpendicular direction of the observation domain, where the origin of the radial direction is the focal point of the laser. We have obtained a displacement distributions that are different among the radial and the circumferential components, which we will discuss in this article.

2. Methodological details

2.1. Experimental setup for observation

The sample consists of 500-nm fluorescent dye-doped polystyrene particles in water at the concentration of $3.8 \times 10^9$ particles/mL contained in the 86-μm thick space as shown in Fig. 1.
The nanoparticles coated with fluorescent dye and they are observed by the inverted microscope with objective lens of ×60 with N.A.0.7 mounted with EMCCD camera, which captures the dynamics of the nanoparticles at a frame rate of 38.1 fps and an exposure time of 25 ms. The trapping optical force originates from the 1064-nm near-infrared laser beam of Nd:YVO₄. The laser power was adjusted by the angle of a half wave plate in front of the polarizing beam splitter.

2.2. Analysis of the movie data
After extraction of trajectory data set from the microscopy movie data in the steady state after the transient period of particles attraction based on the algorithm of Ref. [5], the displacements per each time interval of the camera frames are transformed from $xy$ components to radial $\Delta r_r$ and its tangential, i.e., circumferential components $\Delta r_t$ with an origin defined as the laser spot:

$$\Delta r_r = \Delta r_x \cos \theta + \Delta r_y \sin \theta, \quad (1)$$

$$\Delta r_t = -\Delta r_x \sin \theta + \Delta r_y \cos \theta, \quad (2)$$

where $\Delta r_x$ and $\Delta r_y$ are the displacements in the $x$ and $y$ direction of the camera system, and $\theta$ is the angle comprised of the particle position and the $x$ axis:

$$\theta = \sin^{-1} \left( \frac{y_p - y_o}{\sqrt{(x_p - x_o)^2 + (y_p - y_o)^2}} \right), \quad (3)$$

where $(x_p, y_p)$ and $(x_o, y_o)$ are the positions of the particle and laser spot, respectively.

![Figure 1](image_url)

**Figure 1.** Schematic diagram of the measurement configuration of the sample dispersion. The observation is in the $xy$ plane. The displacement distribution is evaluated in terms of ($\Delta r_r$, $\Delta r_t$) to take into account the effect of laser on the nanoparticle dynamics.

3. Results and Discussion
The laser induces a force field that is basically symmetric in the circumferential direction. There is a trapping effect depending on the distance from the focal point. In other words, the direct influence of the optical force is basically limited in the radial direction. Nevertheless, there is a confinement effect of the cluster in the circumferential direction. The displacement distribution
Figure 2. Radial component of the displacement distribution, where the time span for the displacements is 0.026 s: (a) overall distributions including the rare events, and (b) distributions around small displacements. The legends show the laser power to trap the nanoparticles.

Figure 3. Circumferential component of the displacement distribution, where the time span for the displacements is 0.026 s: (a) overall distributions including the rare events, and (b) distributions around small displacements. The legends show the laser power to trap the nanoparticles.

is sharply peaked around zero regardless of the radial (cf. Figs. 2) and circumferential (cf. Figs. 3) directions or laser power.

However, there is a difference in the displacement distribution for the radial and circumferential directions as shown in Figs. 2(b) and 3(b). Whereas the radial displacements less than 0.3 µm is simply more sharply peaked at zero for larger laser powers, the distributions of
circumferential displacements exhibit non-monotonicity for the larger laser powers. If the particle position distribution is skewed from the circular symmetry to be oval or prolate shapes, it would affect both of the radial and circumferential components based on Eqs. 2 and 3. Therefore, this is attributed to the difference in the radial and circumferential confinement effects.

Furthermore, substantially large displacements appear beyond certain value of the laser power as shown in Figs. 2(a) and 3(a). The clear dependence of the laser power is at least partly attributed to the effect of the scattering force. The scattering force is non-conservative type of the force acting on the nanoparticles. In other words, the nanoparticles steadily receives momentum from the laser in the incident direction (cf. Fig. 1). This qualitative difference from the pure conservative force field can induce the triggered stochastic motion of the nanoparticles. Although the detailed quantitative determination of the force field itself is beyond the scope of this article, it is worth noting that the NA of the lens is not so high nor the particle size is not so small to suppress the scattering force in this experiment. Even though the scattering force from the laser is independent of time, combination with the incessant Brownian motion with direct and indirect (i.e., hydrodynamic) interactions between the nearby nanoparticles results in the marked stochastic motion.

4. Concluding remarks
The nanoparticles trapped by the laser field forms clusters at the solid-liquid interfaces. The constituent nanoparticles experience different confinement effects in the radial and circumferential direction defined by the origin of laser spot and substrate plane. We analyzed the microscopy movie data to evaluate the anisotropy in the Brownian dynamics, and determined the existence of anisotropy in the dynamics. We have obtained counter-intuitive results that the non-monotonicity of the displacement distribution is more pronounced for the circumferential direction compared to the radial direction. This kind of anisotropy might affect the collective structure formation in the crystallization process depending on the combination with other physical conditions and materials.

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