Detection of optical force due to multiphoton absorption

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Abstract. Resonant optical manipulation using absorption force so far has been based on the linear (one-photon) absorption by target material, while optical forces due to multiphoton absorption have not been much investigated. As multiphoton absorption obeys different selection rule from that of one-photon absorption, and also shows non-linear dependence on light intensity, a larger variety of photo-mechanical responses of small particles can be expected by using multiphoton absorption force. In this study, we focused femtosecond laser pulses to a single polymer microparticle containing fluorescence dyes to exert multiphoton absorption force on the particle. We successfully observed the three-dimensional motion of the photo-irradiated microparticle due to the multiphoton absorption.

1. Introduction
Radiation force arising from the exchange of momentum between a small particle and photons can be divided into three terms: gradient, scattering, and absorption forces. The gradient force push the particle toward the spatial gradient of light field, while the scattering and absorption forces direct the particle along the propagation direction of light.[1][2] Among the three forces, the gradient force is usually utilized for the trapping and manipulation of small particles, which is well known as optical tweezer.[3][4][5] On the other hand, the scattering and absorption forces are useful for the transportation of small particles over a longer distance.[6]

In optical micro-manipulation, a laser beam the wavelength of which is not in resonance with target particles is usually employed as manipulation light to avoid photodamage of the particles due to photoabsorption, where the mechanical interaction between photons and particles is not usually so large and relatively high (MW/cm-1) light intensity is required. The momentum of photon is, however, resonantly transferred via the allowed optical transition; strong absorption force can be expected by tuning the wavelength of manipulation light to the peak absorption band of target particles and, this strong absorption force can be used for selective transportation of nanomaterials.[6]

Optical micro-manipulation using absorption force so far has been utilized linear one-photon absorption by target material. On the other hand, optical forces due to multiphoton absorption have not been well investigated. As the multiphoton absorption obeys different selection rule from that of the one-photon absorption, and shows non-linear dependence on light intensity, we can expect a larger variety of photo-mechanical responses of small particles by using the multiphoton absorption. In this study, we aimed to induce and detect the mechanical motion of polymer microparticles due to multiphoton absorption.
2. Experimental Section

2.1. Preparation of sample
Small particles of polystyrene (PS) with fluorescent dyes (perylene and pyrene) were prepared as specimens. Perylene and pyrene were used as two-photon and three-photon absorbers, respectively. 500-nm sized (in diameter) PS particles (Polyscience, Inc.) were used as received. The particles were immersed in ethanol (Wako, 053-06531) solutions of each fluorescent dye. The stock solutions were kept in a dark area in an experimental room for 24 h, leading to the inclusion of each fluorescent dye into the PS particles with ethanol molecules by swelling. The particles containing the dye molecules were separated from the solution by centrifugation, then dispersed into ultrapure water. A drop of the colloidal suspension of the PS particles was injected into a 30-μL sample chamber consisting of two well-cleaned coverslips (Matsunami, 24 × 32 × 0.17 mm) and a 300 μm thick silicon-rubber sheet with a hole.

Fig.1. Structures of (a) perylene and (c) pyrene. (b) Absorption (red line) and fluorescence (blue line) spectra of perylene in benzene. (d) Absorption (red line) and fluorescence (blue line) spectra of pyrene in hexane.

2.2. Optical setup
Each femtosecond (typically 70 fs time duration) light pulse from a mode-locked Ti:sapphire laser (Tsunami, Spectra Physics) operating at a wavelength of 800 nm and 80-MHz repetition was divided into two pulses by a half beam-splitter. A single PS particle is photo-irradiated with a pair of femtosecond light pulses with time-interval, Δt, under a tightly focused configuration by a high numerical aperture (NA) objective (N.A. 1.30/ x100, oil immersion, Olympus). These two successive light pulses were used for trapping the single polymer beads and inducing multiphoton excitation of the dyes in the beads. The relative time delay between the two light pulses, Δt, was controlled by using a stepping-motor-controlled optical delay stage. By controlling Δt, we could change multi-photon absorption strength under the same incident laser power because N-photon absorption strength is in proportion to the Nth power of light intensity.

Fig. 2. Optical setup for inducing and detecting multiphoton absorption force.
2.3. Estimation of the Axial Position

For three-dimensional tracking of single fluorescent particles, we introduced a cylindrical lens into the detection optics to give the imaging system astigmatism.[7] Figure 3(a) shows fluorescence images of a single PS bead at 5 different z-positions obtained with the astigmatism imaging system. These fluorescence images clearly demonstrate that the shape (aspect ratio) of a PS bead are depending of its Z-position. We obtained fluorescence images of the PS bead while slightly changing the Z-position by using a piezo-electric stage. For each fluorescence image the X- and Y-widths are computed by 2D Gaussian fitting and the X- and Y-widths were plotted as a function of the Z-position (Figure 3(b)). The plots were curve-fitted by using the following equation (1) (red and blue solid lines in Figure 3(b)).[8] By using the plot, we determined the Z-position of a trapped PS particle.

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\sigma(z) = \sigma_0 \sqrt{1 + \left(\frac{z - z_0}{d}\right)^2 + A \left(\frac{z - z_0}{d}\right)^3 + B \left(\frac{z - z_0}{d}\right)^4}
\]  

3. Result

Under the condition that \( \Delta t \) was larger than the pulse duration, the particle was optically trapped by the gradient force and slight multiphoton absorption took place by individual light pulses. The number of photons absorbed via multiphoton process increased with decreasing \( \Delta t \) up to the complete overlap of the pulse pair, leading to the increase in fluoresce intensity and change of fluorescence image of the particle due to the astigmatism. By analyzing the fluorescence image of the particles, we confirmed...
that the particles were pushed in the direction of light propagation and the displacement increased with decreasing $\Delta t$ up to $\Delta t = 0$ fs. Furthermore, by increasing $\Delta t$ from $\Delta t = 0$ fs to the initial delay time, the trapping position of the particles returned to the original position. The particles with perylene (two-photon absorber) and pyrene (three-photon absorber) showed similar behaviors. The displacement caused by the multiphoton absorption was in the range from 50 - 200 nm at an input laser power of ca. 1 mW. It should be noted that the displacement varied particle by particle. This is ascribed to the heterogeneity in the number of dyes incorporated in a PS particle as well as the size of the PS particle. For quantitatively evaluating the multiphoton absorption force, detailed experimental parameters such as light intensity distribution at the focus, trapping stiffness of the gradient force, the number of dye molecules in a trapped PS bead, multiphoton absorption cross-section, etc. We are still devoting our effort to obtain such parameters for quantitative estimation of the multiphoton absorption force and the result will be published elsewhere in near future.

We thus demonstrated in the present study the mechanical motion of small particle due to multiphoton absorption force. As multiphoton absorption dominantly takes place only in the focal spot of laser beam under a tight focusing configuration, we can expect spatially selective micro-manipulation by the multiphoton absorption force. We hence foresee a larger variety of photo-mechanical responses of small particles by utilizing multiphoton (non-linear) photo-response.

References

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