[Introduction] A tightly focused laser beam can be used for optical trapping of small objects ranging in size from micro- to nano-scale [1]. When the laser is resonant to the transition between ground and excited electronic states of the objects, radiation force exerted on the objects is enhanced through resonantly enhanced induced polarization. Previously, one of the authors (TK) theoretically calculated the radiation force exerted on the nano-objects including the nonlinear optical effects by taking into account the strong laser intensity required to trap the nano-objects. As a result, it was proposed that the nonlinear optical effects affect the trapping behavior drastically [2]. In particular, the resonance optical trapping with intense blue-detuned laser was considered to give a deeper optical potential than that with red-detuned laser because of an inversion population between ground and excited electronic states [2], although blue-detuned laser does not give favorable condition according to the calculation based on linear optical response [3].

In order to prove our theoretical proposal, we have experimentally demonstrated the resonance optical trapping of individual dye-doped polystyrene nanoparticles with both blue- and red-detuned lasers. As most of the experimental reports [4-6] show the results without operating both lasers because of experimental difficulties, our experiment is the first demonstration of the resonance optical trapping by directly comparing the results by the blue- and red-detuned lasers.

[Experimental] As the objects, we use dye-doped polystyrene nanoparticles suspended in water and bare one (Polyscience) which both diameters are 500 nm. The absorption and emission maximum wavelengths of the dye-doped polystyrene nanoparticles are 524 nm and 548 nm, respectively. As the blue- and red- detuned lasers, we employ 515 nm and 532 nm lasers, respectively. Optical trapping system for blue-detuned laser is constructed on a microscope (Nikon Eclipse Ti) with CCD camera by introducing the laser through oil immersion objective lens (x100, NA 1.4, Olympus). For the red-detuned laser, we use another microscope (Olympus IX-71) with another CCD camera by using the same objective lens. The beam diameters before the objective lens are ~5 mm, and focal height from the bottom of the glass substrate is 50 μm. For confirming the enhancement of the optical trapping, we employ an immobilization time measurement of the individual nanoparticles trapped at the focal spot. Compared with other measurements such as optical stiffness and the number of trapped particles, the immobilization time may give more reliable information on the change of shallow optical potential.
Fig. 1 Histograms of immobilization time measured for 100 trapping events. The laser powers after the objective lens for blue- and red-detuned lasers are 20 mW and 15 mW, respectively. (a) and (b) are blue-detuned laser trapping of dye-doped polystyrene nanoparticles and bare ones, respectively. (c) and (d) are that for red-detuned laser trapping.

**[Results and Discussion]** As shown in Fig.1, for blue-detuned laser we have observed that the immobilization time of dye-doped polystyrene nanoparticles becomes longer than that of bare one, which cannot be explained based on linear optical response. Furthermore, by comparing the resonance optical trapping with blue- and red-detuned lasers, we have first directly found that blue-detuned laser gives longer immobilization time compared with red-detuned laser, and we consider this is originated from the inversion population according to our theoretical proposal. Note that we obtained the similar histograms for bare polystyrene nanoparticles (Figs. 1 (b) and (d)) by adjusting the laser powers and used them as a standard for comparing the results of Figs. 1 (a) and (c), because we cannot have exactly the same focusing condition for both lasers due to experimental difficulties. The present demonstration strongly supports our theoretical proposal on resonance optical trapping based on nonlinear optical response, and we plan to confirm the inversion population more directly by transient absorption spectroscopy.