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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 is significantly greater than that of the bare nanobeads of the same dimension. In striking contrast, the confinement of the nanobeads of the two types was comparable when a continuous-wave laser of the same wavelength and power was employed. The number of confined dye-doped nanobeads increased nonlinearly with the power of the pulsed laser; this dependence was fitted satisfactorily with a second-order polynomial. Supported by theoretical analysis, we attribute the enhanced confinement of doped nanobeads in part to an increased effective refractive index resulting from two-photon resonance between the optical field of the laser and the dopant of the nanobead. We envisage that our findings would evoke applications that benefit from controlled confinement or aggregation of nanomaterials with the employment of near-infrared pulsed lasers.

Keywords: optical confinement, two-photon resonance, nanoparticles, doping, pulsed laser

(Some figures may appear in colour only in the online journal)

1. Introduction

Since its first conception and realization, optical trapping of microscopic objects in a solution with a highly focused laser beam has emerged as a common practice to manipulate particles of micrometer size [1], and has found applications in various disciplines ranging from physics [2–5] and chemistry [6] to biology [7–10]. The optical manipulation of nanometer-sized particles with a great efficiency and stability nevertheless remains a challenge, and is demonstrated almost exclusively on particles made of noble metals or semiconductors [11–13]. Towards this end, various approaches have been attempted, and have led to the discovery of fascinating phenomena. For example, a pulsed laser has been employed to trap Rayleigh particles [13], and to control the dimension and crystalline property of nanodeposits [14]. Some workers employed a pulsed laser about 100 femtoseconds (fs) in duration to manipulate Au nanoparticles in a solution, and observed a split optical potential and two distinct traps in the axial directions [15]. We have reported an alternative ejection of polystyrene nanobeads

(diameter 50 nm) from the laser focus in directions perpendicular to the polarization of the optical field of an intense fs-pulsed laser [16], and suggested that the ejection dynamics were governed mainly by the pulse-width, or the impulsive peak power, of the laser [17]. These results collectively indicate that the strong electric field associated with the fs-pulsed laser can induce nonlinear optical processes [15], or exert an intense repulsive force [16, 17], on the nanoparticles located within the laser focus.

In addition to the intense optical field associated with the ultrashort-pulsed laser, optical resonance represents another distinct mechanism to facilitate optical manipulation [18–22]. We have reported an enhanced trapping of dye-doped polystyrene nanobeads with a combined application of one near-infrared (NIR) and one visible continuous-wave (cw) lasers with the wavelength of the latter falling within the absorption band of the dopant, and suggested that single-photon resonance was responsible for the enhanced trapping [20]. This notion is supported experimentally with an observation of a seven-fold increase in the diffusion time of nanobeads after introducing a second laser that is resonant with the dopants [20], and conforms also to the result of theoretical analysis [18, 22]. Enhanced trapping mediated with optical resonance was formulated theoretically, and realized with only one cw laser with its wavelength falling within the red shoulder of the absorption band of the dopant [21].

A multiphoton optical process has recently been proposed to be an alternative route to facilitate optical trapping, and theoretical analysis therein particularly indicated that two-photon absorption might extend the capability of optical manipulation [23]. Despite being an intriguing idea, this hypothesis has never been verified experimentally. Herein, we investigate a relatively unexplored area of optical manipulation—optical confinement of numerous nanoparticles. Unlike most classical work that addressed mostly enhanced trapping of one particle, we report a strategy to enhance the confinement of numerous nanoparticles. In particular, our work concerns on an increase in the number of particles confined near the focus rather than a greater stiffness of the optical trap. As the intensity of scattering light is proportional to the number of particles [24], we accordingly employed light scattered from particles that were held at the laser focus as an indicator to determine the number of confined particles, and thus the ability of optical confinement.

We characterized the confinement of dielectric nanobeads (diameter 100 nm) with or without doping of dye molecules (absorption maximum near 505 nm) with a pulsed NIR laser (λ = 1064 nm) about 10 picoseconds (ps) in duration. We show that the confinement of dye-doped nanobeads was significantly enhanced relative to that of undoped ones, and the number of doped nanobeads held by the ps-pulsed laser increased nonlinearly with the power of the pulsed laser. Distinct from our previous work that employed a fs-pulsed laser and showed an alternative ejection of particles from the laser focus [16, 17], the employment of a laser of longer pulse-duration (and hence a smaller peak intensity) in this work resulted in a stable confinement of nanoparticles at the laser focus. Supported with theoretical analysis, we suggest that two-photon resonance between the dopant and the ps-pulsed NIR laser mediated an increase in the effective refractive index of the doped nanobeads, thereby accounting in part for the enhanced confinement of nanoparticles. Given the unique advantages rendered by the employment of NIR ps-pulsed lasers such as decreased photodamage and photobleaching of the samples, and greater tolerance to the residual scattering of light especially in a turbulent medium, we envisage that our novel approach should find niche applications that benefit from these attractive features.

2. Methods

2.1. Reagents

Polystyrene nanobeads (Polybead, diameter 100 nm, 2.64% solids; Polysciences) and dye-doped nanobeads (TransFluoSpheres—carboxylate-modified, diameter 100 nm, 2% solids; Invitrogen) were purchased. To minimize the effect of water being heated by the excitation of the overtone of the O–H stretching mode at 1064 nm, heavy water (D_2O, 99.9 atom % D; Aldrich) was used throughout this work.

2.2. Preparation of samples

The sample was prepared on suspension of nanobeads of the designated type in D_2O to a particle density 3.82×10^{13} per ml. A sample cell was assembled with a silicon chamber (diameter 2 cm, thickness 1 mm; EMS) sandwiched between two coverslips (EMS), and was filled with the suspended solution of nanobeads before experiments.

2.3. Excitation and emission spectra of dopants

According to the vendor's information [25], the dopant of our doped nanoparticles comprised derivatives of 4,4-difluoro-4-bora-3a,4a-diaza-s-indacene (BODIPY). Since detailed information about the absorption spectral of the dopant was not disclosed, we measured the excitation spectrum of dye-doped nanobeads suspended in a solution. As shown in figure 1(*a*), the excitation spectrum (blue; λ_{Em} = 561 nm) of the dopant exhibited a peak at 505 nm, and tails extending beyond 450 and 550 nm on the blue and red sides, respectively. As indicated, the half wavelength ($\lambda_{Laser}/2$ = 532 nm; black arrow in figure 1(*a*)) of the ps-pulsed NIR laser employed in this work fell within the absorption band of the dopants. Two-photon resonance is hence possible; indeed, this deduction was verified with the generation of two-photon excited fluorescence after illumination of the solution of doped nanobeads with the pulsed laser.

2.4. Optical setup

The apparatus was modified from a setup that has been described before [16]; figure 1(*b*) displays a schematic of the setup. A pulsed laser (1064 nm, FWHM 7 ps, 76 MHz; PicoTran; High-Q Laser) or a cw laser (1064 nm; Nd:YVO_4; Spectra Physics) was employed in this work. The laser beam

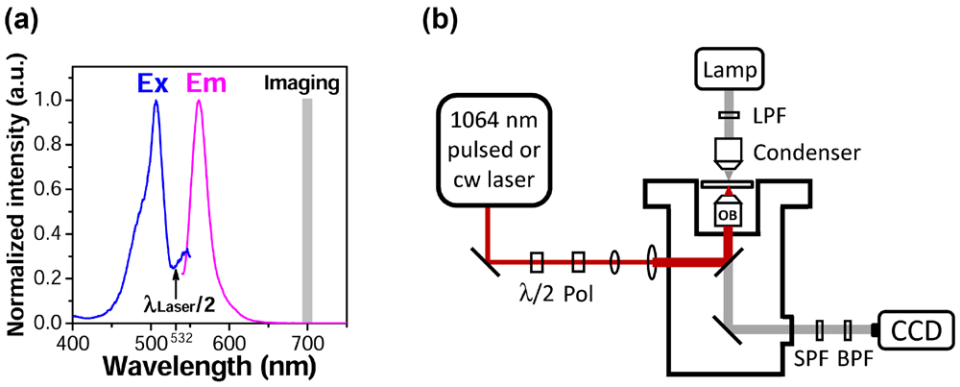

Figure 1. (*a*) Excitation (blue; $\lambda_{Em.}$ = 561 nm) and emission (pink; $\lambda_{Ex.}$ = 532 nm) spectra measured from a solution of dye-doped nanobeads employed in this study. The grey bar indicates the pass band of the filter employed to retain only the scattering of light from the condenser, while blocking incidence of either the two-photon induced emission, or the scattering of light from the near-infrared laser, to the CCD camera. (*b*) Experimental setup. *λ/2*: half-wave plate; Pol: polarizer; LPF: long-pass filter; SPF: short-pass filter; BPF: band-pass filter; CCD: charge-coupled device; OB: objective lens.

was collimated and directed to an inverted optical microscope (Eclipse Ti; Nikon), and was focused onto the sample with an objective lens (60×, NA 0.95; PlanApo; Nikon). The focus of the laser beam was set 100 μm above the upper surface of the bottom coverslip of the sample cell. We determined the ability of confinement from the intensity of light scattered from nanobeads using a setup modified from that reported before [16]. In brief, the sample was illuminated with a halogen lamp through a microscope condenser (NA 0.52); images of scattered light resulting from illumination of the condenser lamp were recorded with a camera (CV-A55IR E from JAI, or Wat-902 H-1/2 from WATEC) operated at 30 interlaced frames per second. To prevent the dopants from being excited by the illumination of the lamp, only light between 630 and 750 nm was allowed to pass through the condenser with a long-pass filter (LP01-633; Semrock). A short-pass filter (FES900; Thorlabs) and a band-pass filter (FB700-10; Thorlabs) were also placed before the camera to block either the Rayleigh scattering produced from illumination of the NIR laser, or fluorescence emission resulting from two-photon excitation. With such a setup, all light detected by the CCD camera is exclusively the Rayleigh scattering resulting from illumination of the condenser lamp (rather than that of the laser, or the fluorescence emission) on the nanoparticles that were confined near the focus (the pass-window was highlighted with a grey band in figure 1(*a*)). As described, the intensity of scattering light is proportional to the number of particles [24]. Light scattered from particles was hence employed as an indicator to determine the number of particles confined at the laser focus, and thus the ability of optical confinement. The acquired images were digitized and analyzed with commercial software (ImageJ).

3. Results

We first attempted to manipulate nanobeads with or without dopants using a ps-pulsed laser. The image of scattered light recorded on the confinement of nanobeads of either type shows a discernible bright spot relative to the dark background, indicating that numerous nanoparticles were confined at the laser focus (figure 2(*a*)). The cross-sectional profiles of the images show that the intensity is several times greater for the confinement of doped nanobeads relative to their bare counterparts (figure 2(*b*)). We evaluated also the stability of the confinement, and found that the intensity of scattering light increased immediately after the laser was introduced to the solution of doped nanobeads and remained at a stable level thereafter; in comparison, the increase of the intensity observed on the confinement of bare beads was less profound after introduction of the laser (figure 2(*c*)). As described, the intensity of scattering light can be regarded as an indicator of the number of particles confined by the laser; these collective results hence show unambiguously that the ps-pulsed laser confined more doped nanobeads than undoped ones. The result indicates that the doping of dye molecules to the nanobeads might play a key role in the enhanced confinement of doped nanobeads. Furthermore, as the half-wavelength of the pulsed laser employed in this work fell within the absorption band of the dopants and the pulsed laser possessed a large peak power, we thus attributed the distinctly enhanced confinement of doped nanobeads to two-photon resonance between the ps-pulsed NIR laser and the dopants. We return to this point in the subsequent sections.

As the photon density of a cw laser is smaller than that of a pulsed laser of the same average power by several orders of magnitude, a cw laser of a moderate power is unlikely to induce two-photon absorption. As a negative control, we employed a cw laser of the same wavelength and average power as that of the ps-pulsed laser employed in the preceding experiments to confine nanobeads. The images obtained on the confinement of doped and undoped nanobeads show that there was a discernible but dim spot in either image (figure 3(*a*)). As illustrated in the cross-sectional profiles of the two images, the intensities of the two spots are comparable indicating that the number of doped nanobeads was comparable with that of bare nanobeads (figure 3(*b*)). Furthermore, the temporal intensity fluctuated dynamically

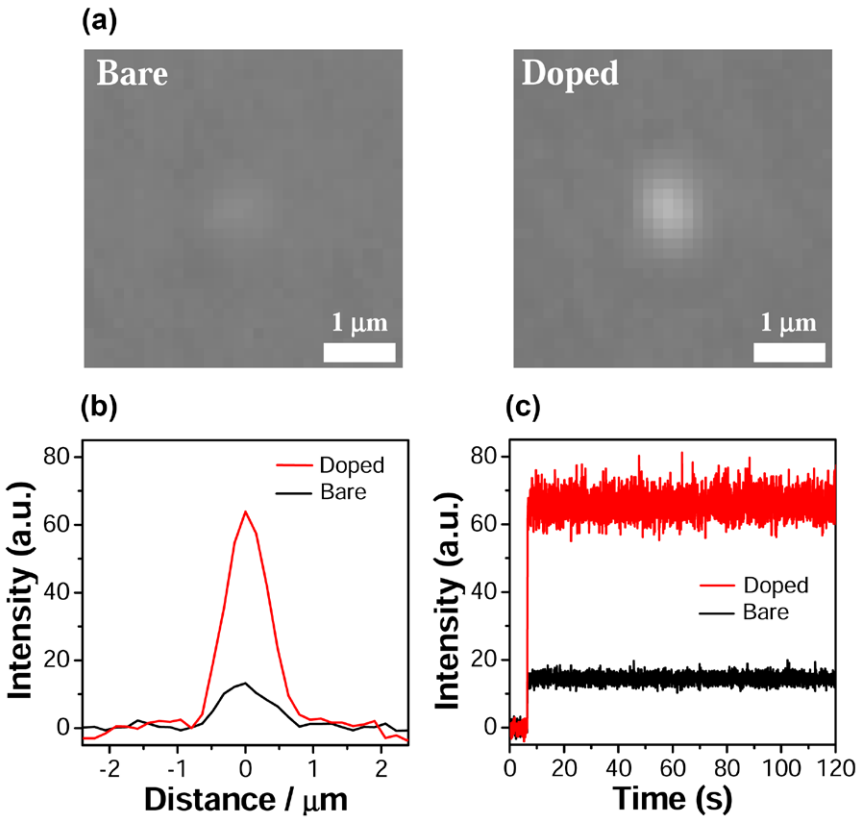

Figure 2. Optical confinement of nanobeads using a ps-pulsed laser. (*a*) Bright-field images of optically confined nanobeads dispersed in heavy water. (*b*) Cross-sectional profiles of the images. (*c*) Temporal intensity.

when a cw laser was employed (figure 3(*c*)). In comparison with the results displayed in figure 2, it became clear that the result obtained with the cw laser is distinct from that with a pulsed laser of the same wavelength and power. Particularly, the present result indicates that, when a cw laser was employed, the doping of dye molecules into the nanobeads had little effect on optical confinement of these nanobeads. As described, a cw laser was unlikely to induce absorption of the dopant through a two-photon process; this negative result hence conforms to our deduction obtained from the preceding experiments.

To further consolidate our deduction that two-photon resonance might contribute to the enhanced confinement of doped nanobeads, we proceeded to examine how optical confinement depended on laser power. When the pulsed laser was employed, the number of nanobeads of either type confined at the laser focus increased with the laser power; particularly, the increase was much steeper for the confinement of doped nanobeads relative to that of the undoped ones, exhibiting a nonlinear dependence that was satisfactorily described with a second-order polynomial, whereas the result for the undoped nanobeads showed a linear increase with laser power (figure 4(*a*)). As a comparison, we employed a cw laser and performed the experiments in the same manner; in sharp contrast with the preceding observation made with the ps-pulsed laser, the confinement was comparable in magnitude and increased linearly with the laser power for nanobeads of both types (figure 4(*b*)).

4. Discussion

We have demonstrated a novel strategy to enhance optical confinement of dielectric nanobeads. We show that the ps-pulsed NIR laser confined more dye-doped nanobeads than bare ones of the same size (figure 2). The ability to confine doped nanobeads exhibited a nonlinear dependence on laser power, and the dependence was fit in good agreement with a second-order polynomial, whereas the confinement of bare nanobeads exhibited a linear dependence (figure 4(*a*)). In contrast, the ability to confine doped nanobeads or bare ones was comparable when the cw NIR laser was employed (figure 3), and the ability to confine nanobeads of either type depended linearly on laser power (figure 4(*b*)). With these collective results, we suggest that a nonlinear interaction, particularly two-photon resonance between the dopant and the pulsed laser, might be responsible for the enhanced confinement of doped nanobeads by the ps-pulsed laser.

To rationalize our observation, we adapted the classical theory of optical trapping in the Rayleigh regime, and evaluated the relative contribution of the trapping force and the pushing force. In this regime, the trapping force acting on the particle is a gradient force and the pushing force comprises mainly two terms, the scattering force and the absorption force. Besides, the gradient force is a function of the real part of the complex polarizability of the particle whereas the scattering force scales with the polarizability squared and the absorption force depends on the imaginary part of the complex polarizability [21].

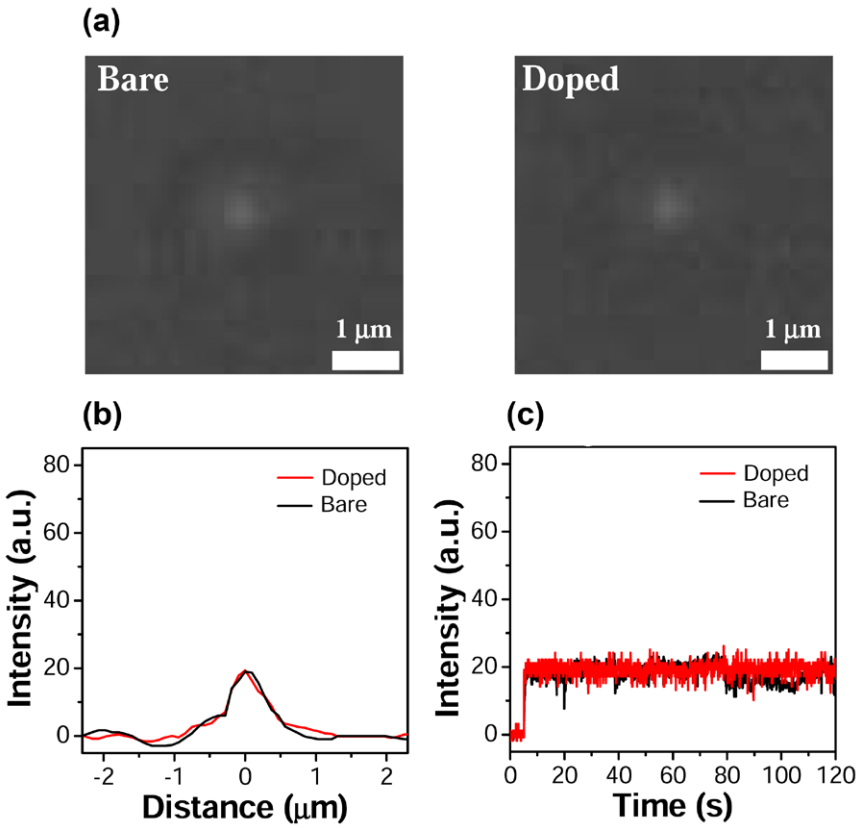

Figure 3. Optical confinement of nanobeads using a continuous-wave laser. (*a*) Bright-field images of optically confined nanobeads dispersed in heavy water. (*b*) Cross-sectional profiles of the images. (*c*) Temporal intensity.

We first argue that the gradient force is much greater than the scattering force for small particles. As described, the gradient force and the scattering force scale with the real part of the complex polarizability of the particle and the polarizability squared, respectively. As the polarizability scales with the volume of the particle, the scattering force is hence insignificant relative to the gradient force for small particles (i.e. in the Rayleigh regime) [26]. In this work, the dimension of nanobeads (radius = 50 nm) is much smaller than the wavelength of the laser (λ = 1064 nm) employed to confine these nanobeads. Accordingly, the contribution of the scattering force acting on the nanobeads used in this work is insignificant relative to that of the gradient force.

We then argue that it is possible to choose the wavelength of the laser such that the gradient force is greater than the absorption force. As already described, the gradient force and the absorption force scale with the real and the imaginary part of the complex polarizability of the particle, respectively. Moreover, both the real and the imaginary parts of the polarizability depend on the frequency of the optical field near resonance, and exhibit a dispersive and a Lorentzian profile, respectively. It has been reported that the increase of the real part of the polarizability could exceed that of the imaginary part on the low-frequency side of the resonance [21]. Accordingly, the gradient force can be greater than the absorption force as the laser is chosen such that the wavelength of laser is near and above the maximum of the absorption band of the sphere. This is the origin of enhanced trapping mediated by optical resonance.

Although the above conclusion is deduced within the context of single-photon resonance, we assume that this conclusion applies also to the case of two-photon resonance. As described, a condition of two-photon resonance was fulfilled between the optical field of the ps-pulsed NIR laser and the dopant, which was verified by the generation of two-photon excited emission (data not shown). Although the detail about the two-photon absorption spectrum of the dopant is unknown, the peak of the two-photon absorption spectrum tends to become blue-shifted relative to that predicted by simply assuming twice the peak of the single-photon absorption spectrum [27]. Moreover, as the half-wavelength of our laser ($\lambda_{\text{Laser}}/2$ = 532 nm) is significantly greater than the peak of the excitation spectrum determined with single-photon excited emission ($\lambda_{\text{Ex.}}$ = 505 nm; see also figure 1(*a*)), it is hence likely that the wavelength of our laser is located on the low-frequency side of the two-photon absorption spectrum of the dopants. Accordingly, the gradient force acting on the doped nanobeads also overweighs the absorption force.

With these deductions taken together, we conclude that the contribution of the pushing (scattering and absorption) force that acted on the doped nanobeads is less significant relative to that of the trapping (gradient) force in our experiments. The preceding discussion rationalizes the confinement of doped nanobeads with a ps-pulsed 1064-nm laser, and, more importantly, supports that two-photon resonance is a contributing factor to enhancing the confinement of dye-doped nanobeads relative to bare nanobeads.

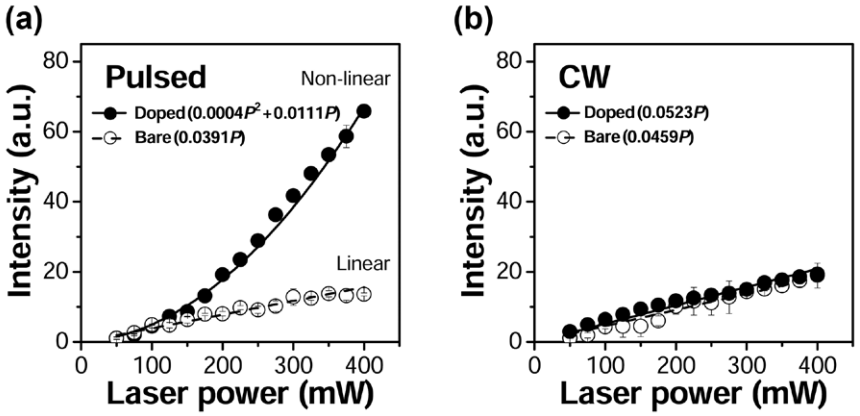

Figure 4. Intensity of the scattered light as a function of the average laser power (solid circles: dye-doped beads; empty circles: bare beads). (*a*) and (*b*) were obtained using pulsed and cw lasers, respectively. The raw data were fitted with a linear function or a second-order polynomial; the results obtained from the best fit are displayed as insets (*P* denotes the laser power).

We proceed to explore the mechanism of the enhanced optical confinement in more details below. As described, the doped nanobeads employed in this work comprise dielectric materials (polystyrene) and dopants (dye molecules). The dielectric material is dominant but is resonant with neither the fundamental nor the second harmonic of the optical frequency of the laser, whereas the less abundant dopant is resonant with the second harmonic of the laser [28]. The effective refractive index of a weakly absorbing sphere ($\tilde{n}$) can be defined as [29],

$$\tilde{n} = \sqrt{1+\tilde{\chi}_{\text{eff}}} \tag{1}$$

where $\tilde{\chi}_{\text{eff}}$ is the effective complex susceptibility of the weakly absorbing sphere. Under the two-photon absorption transition, the effective susceptibility can be expressed as,

$$\tilde{\chi}_{\text{eff}} = \tilde{\chi}^{(1)} + \frac{3}{4}\tilde{\chi}^{(3)}|\vec{E}|^2 \tag{2}$$

where $\tilde{\chi}^{(1)}$ is the linear susceptibility associated with the linear refraction and absorption, and $\tilde{\chi}^{(3)}$ is the third-order nonlinear susceptibility describing the nonlinear refraction and absorption. Under our experimental condition, there is no single-photon resonance at the fundamental frequency of the laser; $\tilde{\chi}^{(1)}$ is hence a real quantity at that frequency. Accordingly,

$$\tilde{n} = \sqrt{1+\chi^{(1)}+\frac{3}{4}\tilde{\chi}^{(3)}|\vec{E}|^2} \approx \sqrt{1+\chi^{(1)}}\left(1+\frac{3}{8}\cdot\frac{\tilde{\chi}^{(3)}|\vec{E}|^2}{1+\chi^{(1)}}\right) \tag{3}$$

With the linear refractive index of the sphere defined as $n_0 \approx \sqrt{1+\chi^{(1)}}$, the nonlinear refractive index as $\tilde{n}_2 = 3\tilde{\chi}^{(3)}/4\varepsilon_0 c n_0^2$ and the intensity of light as $I = \frac{1}{2}\varepsilon_0 c n_0 |\vec{E}|^2$, the effective refractive index of the weakly absorbing sphere can be approximated as

$$\tilde{n} = n_0 + \tilde{n}_2 I \tag{4}$$

In general, n_0 and $\tilde{n}_2$ are quantities that depend on the frequency of the optical field. As the absorption band of the dopants (centered at 505 nm) is remote from the fundamental frequency of the laser, there is consequently no single-photon resonance at the fundamental frequency of the laser, and n_0 is thus essentially a real quantity; in contrast, $\tilde{n}_2$ is a complex quantity and is appreciable at the fundamental wavelength of the laser as the two-photon resonant condition can be energetically fulfilled (i.e. the half wavelength of the laser falls within the absorption band of the dopant) [28]. Furthermore, the contribution of the nonlinear term ($\tilde{n}_2 I$) to the effective refractive index of the doped nanobeads also becomes appreciable when the intensity of the laser is large, as in the case of a pulsed laser [28].

We proceed to rationalize our observation in more detail. It is plausible to assume that the greater the trapping force the more particles confined by an optical trap. As an approximation, we assume that the ability of optical confinement is proportional to the force exerted on the doped particles. As described, the magnitude of the gradient force responsible for optical trapping is proportional to the gradient of the optical field (∇I; I denotes the intensity of the optical field) multiplied by the real part of the polarizability of the particles ($\text{Re}(\alpha)$; α denotes the polarizability of the particle),

$$F_{\text{gradient}} \propto \nabla I . \text{Re}(\alpha) \tag{5}$$

with the former quantity proportional to the intensity of the laser and the latter quantity to the ratio of the effective refractive index of the particle to that of the surrounding medium [21]. As derived in the preceding paragraph, the effective refractive index of the doped nanobeads comprises a constant term and a term depending linearly on the intensity of the laser (4). Combining these results, our derivation indicates that the dependence of the trapping force on the laser power is describable with a linear term and a quadratic term. We have shown that the dependence of the confinement ability of doped nanobeads is described satisfactorily with a second-order polynomial that comprises a linear term and a quadratic term (figure 4(*a*)). This deduction is thus qualitatively consistent with the experimental results. Our simple argument has hence captured the essential feature of the dependence of the confinement ability on the laser power. A quadratic dependence of the trapping force on the laser intensity has been reported for the trapping of non-resonant Kerr particles, and the quadratic dependence was rationalized with the generation

of a power-dependent refractive index of the Kerr material illuminated with an intense laser [26].

We are not aware of any report on the nonlinear refractive index of dye-doped polystyrene nanobeads. We however note that the nonlinear refractive index of a polystyrene film doped with derivatives of BODIPY is on the order of $10^{-12}\,m^2W^{-1}$ [30]. With the laser power at the range of a few hundred mW, the intensity at the focus of a high numerical-aperture objective lens is on the order of $10^{11}\,W\,m^{-2}$. With these results collectively, the contribution of the nonlinear term ($\tilde{n}_2 I$) to the effective refractive index could be appreciable if an intense laser is employed to confine doped particles under the condition of two-photon resonance. Notably, the conclusion of this simple argument conforms to our observation of an increased ability to optically confine doped nanobeads with the intense NIR pulsed laser.

As described in the introduction, enhanced trapping of doped nanoparticles has been realized with a cw laser through single-photon resonance by some groups including ourselves [20, 21]. We have also demonstrated the employment of a fs-pulsed laser to manipulate dielectric nanoparticles [16, 17]. Our result is nevertheless distinct from these earlier works in several respects. Firstly, unlike the use of a cw laser to trap doped particles and to fulfill the condition of single-photon resonance [20, 21], we employed a pulsed laser with its half-wavelength falling within the absorption band of the dopant; consequently the resonant condition was fulfilled most likely through a two-photon process. The observation of a quadratic dependence of the confinement ability on the power of the laser also strongly supports this notion (figure 4(*a*)). As the cross-section of two-photon absorption is significantly smaller than that of a single-photon counterpart, we did not expect a significant enhancement when the laser power was small. Our result indeed shows that the enhancement was inappreciable before the laser power exceeded 150 mW and increased as the laser power increased (figure 4(*a*)). The result of the curve fitting also supports this notion, as shown from the small coefficient of the quadratic term relative to that of the linear term (0.0004 versus 0.0111). Secondly, unlike the concurrent observation of enhanced trapping and alternative ejection of nanoparticles from a laser focus when a fs-pulsed laser was employed [16, 17], the employment of a ps-pulsed laser in this work resulted in a stable confinement with no evidence of alternative ejection of nanobeads (figure 2). As the ejection of the nanoparticles resulted from the temporal force (a repulsive force) that is inversely proportional to the temporal duration of the pulse [16, 17], it is thus unsurprising that this distinct phenomenon was not observed in this work in which a laser with a pulse of large duration (10 ps versus 100 fs) was employed. The minimized temporal force and hence lack of alternative ejection represents a unique feature of our approach relative to what was observed in confinement with fs lasers, and should be advantageous in applications in which alternative ejection of particles is undesirable. In general, the pushing (scattering and absorption) force could also result in ejection, or stochastic kick, of nanoparticles out of the confinement site. As discussed in the preceding paragraphs, the pushing force is presumably much smaller than the gradient force under our experimental conditions. Its effect is hence omitted in our work.

5. Conclusion

We have explored for the first time a two-photon resonance as a novel means to facilitate optical confinement of dielectric nanobeads. Through the employment of a picosecond-pulsed laser of which the half-wavelength falls within the absorption band of the dopants, we demonstrated an increased ability to confine doped nanobeads relative to their undoped counterparts. We suggest that the enhanced confinement was facilitated in part through an additional term in the effective refractive index of the doped nanobeads resulting from a nonlinear optical interaction between the intense laser and the dopants and through the large peak power of the pulsed laser.

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