Research article

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Proposed method for highly selective resonant optical manipulation using counter-propagating light waves

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Abstract: Optical manipulation using electronic resonance can realize the selective manipulation of nano objects exhibiting quantum mechanical properties by confining electronic systems based on the characteristics of individual objects. This study theoretically proposes a method to actualize selective manipulation based on the resonant optical response. In this method, counter-propagating light waves are used to extract the pure contribution of the resonant optical response in the exerted force by regulating the balance between the two light waves. Furthermore, the selection of nanoparticles with particular resonance levels at room temperature and quantum dots with a particular size in the cryogenic condition is numerically demonstrated. An especially interesting aspect of this method is that it enables the examination of the absorption spectrum of a single nanoparticle by mapping the absorption efficiency to its mechanical motion. The results reveal an unconventional link between optical force technology and nanomaterials science.

Keywords: nano particles; optical force; optical manipulation; resonant optical response.

1 Introduction

According to the electromagnetic theory by Maxwell, light conveys momentum, and if light interacts with a substance, the momentum is transferred to the substance and a mechanical force is exerted. This force is sometimes called optical force, and is of two types: (a) dissipative force, which arises from the transfer of light momentum to a substance by absorption and scattering, and (b) gradient force arising from an electromagnetic interaction between induced polarization and incident light. The dissipative force is useful for transporting particles, and the gradient force is generally used for trapping particles. Optical manipulation has been theoretically proposed and experimentally demonstrated by Ashkin et al. as optical tweezers [1, 2] to trap and manipulate micrometer-scale objects using optical force, and has been employed in various research fields, such as biophysics, physicochemistry, and micromechanical engineering [3–9]. In addition, methods of atom cooling have been established as one of the optical force technologies [10].

Recently, nanometer-scale objects have been of increasing interest for optical manipulation. If the direct trapping and flexible manipulation are possible for nano-objects, such as quantum dots (QDs), nano carbons, and organic molecules, it would be possible to realize highly sensitive sensing, novel nanofabrication technologies, and efficient nanometrology. However, manipulation of individual nanoparticles (NPs) remains challenging, because the optical force is very weak, i.e., it is approximately proportional to the particle volume in the Rayleigh scattering regime. Thus, overcoming the disturbance from the usual environment of NPs is difficult; the movement of solvent molecules or something interferes with the manipulation. To enhance the force, using evanescent field with a steep gradient of electric field has been proposed [11, 12]. Recently, trapping of NPs associated with plasmonic resonance is being extensively studied [13–15]. Highly localized and enhanced electric field arising from the localized surface plasmon resonance near the metallic

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structures induces a strong gradient force, which is sufficient for the trapping of NPs.

Furthermore, in recent years, optical manipulation based on electronically resonant optical response has been proposed to enhance the force [16–22]. Interestingly, using resonant light makes possible the selective manipulation of individual targets with specific physical properties because the optical response of NPs strongly depends on the quantum mechanical properties of individual particles. Moreover, successful NPs trapping and transport using resonant laser light have been reported [23–27]. These studies were based on the condition that the resonance peak of each NP is discrete, and the incident light with specific photon energy can selectively enhance the interaction with the corresponding particles (Figure 1(a)). However, realizing selective operation under general conditions is challenging. In particular, selective manipulation with the resonant effect is difficult under the following conditions: (1) When the resonant bodies are embedded in the mother matrix (similar to a nano-diamond with NV-centers or a dye-doped polymer particle), the magnitude of optical force applied to the whole particle is almost determined by the matrix size. Under this condition, selective manipulation based on the physical properties of the embedded resonant bodies is difficult (Figure 1(b)). (2) The size of the QDs can be sorted by the optical force at the excitonic absorption edges because of the quantum confinement effect. However, if the spectral peak

structures of the targeted QDs are masked by higher energy absorption of the other QDs with lower absorption edge, selective manipulation is difficult (Figure 1(c)).

The objective of this study is to theoretically propose a scheme to actualize the concept of resonant optical manipulation that is not affected by the above-mentioned problems, where the selectivity of optical manipulation based on the resonant optical response is substantially enhanced under general conditions. The proposed scheme extracts the component of the pure resonant contribution and reflects it in the mechanical motion of NPs. For this purpose, the contributions of objects other than the targeted resonance objects are cancelled out using counter-propagating light waves by regulating the balance between the forces using two light waves by tuning their energies, intensities, and polarizations (Figure 1(d)). Counter-propagating light waves were used in a previous study of Ashkin, Ref. [1], for the three-dimensional trapping of targeted small particles using only the dissipative force; numerous similar setups have been tried since then [28, 29]. Recently, Ploschner et al. demonstrated sorting gold nanoparticles according to size using counter-propagating light waves [30]. Using size dependent optical spectra and regulating the intensity balance of the propagating light waves, they could control the force direction according to the size of nanoparticles. Additionally, some studies used thermal fluctuation, thermo-hydrodynamics, or heating to control, depending on size, the optical force on metallic

Figure 1: (a) Schematic of the force spectra of NPs with size-dependent resonance peaks. Incident light of a specific energy can selectively enhance interaction with the corresponding particles, and we can thus selectively transport them. (b) Schematic of force spectra of NPs, where the resonant bodies are embedded in the mother matrix. The optical force applied to the whole particle is almost determined by the matrix size. In this diagram, the exerted force is stronger for the particle with the spectrum represented in red, even if the photon energy is tuned to the peak of the spectrum, which is represented in green. (c) Schematic of force spectra of the QDs of two groups in regions of different sizes. In this case, selective manipulation of the QD group with a higher absorption edge (represented in green) is difficult because the force on the group with a lower absorption edge (represented in red) is stronger at the peak position of the former. (d) Schematic of optical manipulation with counter-propagating light waves.

nanoparticles or its direction based on the use of the counter-propagating light waves [31–33]. In contrast to the above works, counter-propagating light waves in the present work serve a different purpose, i.e., cancelling the non-resonant component in the optical force, which enables the extraction of the pure force component arising from the electronic resonance. This scheme can select particles, including aimed resonance levels, regardless of whether their size is different from, or the same as, those of other particles.

Further, a foremost objective of the proposed method is to measure the optical property constants of a single NP by observing its mechanical motion, which can be realized by extracting the pure component of the optical force arising from the resonant optical response. Hitherto, there have been several trials to measure the optical properties of single nanostructures [34]. Tamaru et al. measured light scattering from individual Ag nanoparticles [35]; furthermore, Okamoto and Imura visualized optical fields in metal nanostructures based on near-field imaging [36]. Recently, elaborate techniques enable accurate dark counting of photons from single metallic nanoparticles, which realize single nanoparticle extinction measurement [37]. Furthermore, the absorption of single particles not associated with scattering can be measured using photothermal imaging techniques [38, 39]. Although these studies have significantly developed single particle spectroscopies, the direct measurement of the resonant absorption process without being mediated by any relaxation processes (including radiative dissipation) remains a challenge. The method by Sandoghdar and colleagues using a balanced detection technique [40, 41] is a powerful tool for observing single-molecule-level absorption. However, extracting pure absorption signals not affected by any scattering component or attenuation of the transmitted intensity would be challenging. Our proposed idea realizes the direct measurement of resonant absorption through the momentum change that is not affected by any energy relaxation processes or chemical reactions. Obtained information by this method would be crucially important for the analysis and design of the interaction strengths between light and single matter states.

In the rest of this paper, we present the numerical investigation of the feasibility of selective manipulation by considering the following three cases: [A] Quantum emitters embedded in a mother matrix at room temperature: in this case, we usually encounter the above-mentioned problem (1), which can be addressed by cancelling the force contributions from the mother matrix using counter-propagating light waves. If the photon energy of one light wave is in the non-resonant region and if the other wave is

tuned to the resonance level, we can select particles with the aimed resonance levels by regulating the intensity balance between the two light waves to cancel out the non-resonant contributions of optical forces. [B] In this investigation, we further demonstrate the measurement of the absorption spectra of a single particle by observing the motion of NPs. In the cases [A] and [B], we assume diamond nano-crystals including nitrogen-vacancy (NV) centers as NPs whose schematic spectra are shown in Figure 1(b). [C] QD assembly: in this investigation, we consider the case in which we selectively transport QDs within a particular absorption line, in a QD ensemble with inhomogeneous lines under the cryogenic condition, similar to that in Ref. [23]. In the cryogenic condition, such as in liquid helium environment, the effects of viscosity/friction and Brownian motion are quite small and, hence, the particles can be easily moved by very weak intensity of laser [23]. Furthermore, the resonance lines of particles become very sharp because of much lower effect of phonons. This significantly enhances the selectivity of the resonance optical manipulation. However, by using a single laser beam, it is difficult to utilize this great opportunity even under such ideal condition if we encounter the problem (2) mentioned above (Figure 1(c)). In this demonstration, we consider the situation where the resonant absorption spectra of targeted QDs are masked by the higher energy absorption of another size group of QDs with the lower absorption edge.

For [A]~[C], the methods for calculating the optical force and the Brownian simulation are provided in the Section 4.

2 Results and discussion

2.1 Selective manipulation of quantum emitters embedded in mother matrix

Here, we consider a scenario in which a single diamond nano-crystal with 90 NV-centers (DNVC) and a pristine diamond nano-crystal (PD) are irradiated by an evanescent field on a glass substrate in water medium (Figure 2(a)). When the nano-diamonds are used in water, they are usually detergent-solubilized to prevent them from aggregating. If two close nanoparticles, including resonance levels, are irradiated by resonant light, interaction between them via the optical force and optical binding effect generally occur [42, 43]. However, we assume the low density of DNVC here and do not explicitly treat such effects to focus our demonstration and discussion on the essence of our idea, which is to avoid additional

complicated phenomena. Furthermore, to avoid the interference effect, the incident light wave 1 is set to the TM mode and the light wave 2 to the TE mode. The incident angle θ_i is set as $\theta_i = 1.1$ rad, to exceed the critical angle 1.09 rad. As the targets, we assume the following parameters. [Diamond]: The diameter of PD is 45 nm, and that of DNVC is 40 nm; the dielectric constant of diamond is 5.77 for infrared region and 5.84 for visible region. [NV-center]: The NV-center is modeled as a simple two-level system. The transition energy is $\hbar\omega_{21} = 2.32$ eV. The dipole moment is $\mu_{21} = 4.0$ Debye, and the dephasing constant is $\gamma_{21} = 20$ meV. In the following demonstrations, we do not consider absorption saturation because our experiment does not show the effect for assumed light intensity (unpublished result.)

The spectra of optical forces for both DNVC and PD are shown in Figure 2(b). In this figure, broken lines represent the forces of only light wave 1 (photon energy is changed from 2.0 to 2.7 eV, and the intensity is 5 MW/cm²). In the case of the present size combination, the force exerted on the PD becomes stronger even if the photon energy is tuned to the resonance of NV-centers in the DNVC as shown in the broken lines in Figure 2(b); namely, the scattering force of PD is larger than the absorbing force arising from the resonance of the NV-center. Therefore, selecting an DNVC with single light-wave irradiation is impossible even if we tune the photon energy to the absorption line of the NV-center. On the other hand, solid lines represent the forces of

the counter-propagating light waves (photon energy of light wave 1 changes as mentioned above, and the photon energy of light wave 2 is 1.16 eV, which is in non-resonant region, and its intensity is 105 MW/cm²). Here, we can realize a scenario where the directions of the optical forces on DNVC and PD are opposite to each other near the resonance of the NV-centers by regulating the intensities and photon energies of the two light waves (see the inset in Figure 2(b)). Aiming at this condition, selective optical manipulation can be performed.

Figure 2(c, d) shows the trajectories of the two particles obtained by Brownian dynamics simulation. We assumed an aqueous environment on the glass substrate at 300 K. The environmental parameters used for this simulation are provided in the Section 4 (Kinetic analysis). Figure 2(c) shows the case of the irradiation of only light wave 1, with the energy being equal to the transition energy of the target. In this case, the force on PD becomes stronger because its particle size is larger. Thus, its transport distance is longer, and sorting the resonant particles thus becomes difficult. On the other hand, Figure 2(d) shows the case of the irradiation of counter-propagating waves when the energies of light wave 1 and light wave 2 are set as 2.32 and 1.16 eV, respectively. As seen in the inset of Figure 2(b), the force on DNVC is positive, whereas that on PD is negative, in this case. Thus, DNVC moves toward positive z and PD moves toward negative z . This further indicates that

Figure 2: (a) Model of optical manipulation of nano-diamond particles with NV-centers by the counter-propagating evanescent waves at the interface between glass and water. The dielectric constants of glass and water are assumed to be 2.25 and 1.77, respectively. The parameters of nano-diamond and NV-centers are described in the text. (b) Optical force spectra of DNVC and PD. The green and red lines represent the forces exerted on DNVC and PD, respectively. The dashed lines represent the forces with only light wave 1, and the solid lines represent the forces with the counter-propagating light waves. (c) Trajectory of two particles for 5 s transport with only light wave 1 (photon energy is 2.32 eV and light intensity is 5 MW/cm²). (d) Trajectory of two particles for 20 s transport with the counter-propagating light waves. Photon energy is 2.32 (1.16) eV and light intensity is 5 (105) MW/cm² for wave 1 (2).

the particles are more strongly bound to a plane by the stronger evanescent field than that in the case with only light wave 1 (see the scale of the vertical axes). Actual experiment revealed that confining the motion of NPs within a limited dimension is effective for observing particle motion in the present scheme using counter-propagating light waves. The results of such experiment using a nano-fiber are planned to be presented elsewhere.

The above demonstration shows a typical case in which selecting the targeted NP is difficult using only a single light wave. The above method (by counter-propagating waves with spectrally distant wavelengths) is effective for the selection of the targeted NPs (DNVC) with a particular resonance line, even if there are many non-resonant NPs, as long as their size is uniform (regardless of whether their size is the same as the resonant NP or not.) However, if the size of non-resonant NPs is dispersed, regulating light intensities for individual non-resonant NPs is impractical. For such a case, using counter-propagating light waves with spectrally near frequencies is effective for selecting the resonant NPs with a particular resonance line. Because the force spectrum of non-resonant NP is always a monotonically increasing function of the light frequency, the force is toward the direction of propagation of a light wave with upper frequency for any non-resonant NP; on the contrary, we can invert the force direction only on resonant NPs because the force spectrum is a decreasing function just above the spectral peak of resonant NPs (see, the green dashed line in Figure 2(b), for example), and we can control two frequencies so that the spectrum has a negative slope between them. Thereafter, NPs with a particular resonance line could be selected. A similar type of operation is demonstrated in subsection C.

2.2 Measurement of absorption spectrum of a single NP

Then, we explain the method to make the motion of a single NP reflect its absorption spectrum by extracting only the force component from resonance using counter-propagating light waves (Figure 3(a)). In this demonstration, a target particle is one DNVC comprising a mother matrix of diamonds without any absorption and embedded 90 NV-centers as absorbers. We measure the absorption spectrum of this particle. First, we maintain a balance between the forces using two light waves below and above the resonance (indicated by blue arrow and yellow arrow, respectively, in the inset of Figure 3(a)) by regulating the

intensities of both light waves. Then, from the intensity ratio of the two light waves, we can estimate the intensity to maintain the balance only for the background contributions (namely, the balance between the scattering forces indicated by the yellow outlined arrow and green outlined arrow) when the frequency of one light wave is in the resonance region using the Clausius-Mosotti equation, as shown in Section 4 (Optical force). If NV-centers exist in the targeted nano-dia, this balance is lost because of the resonant contribution in the force (i.e., the absorbing force), and the DNVC moves. This motion arises solely from the force component because of the resonance indicated by the green filled arrow in Figure 3(a). Therefore, this operation can be used to measure the optical absorption of a single DNVC by measuring the particle transport distance. In the present case, we sweep the frequency of light wave 1 by adjusting the intensity to maintain a balance between the background contributions (scattering forces) by two light waves (indicated by the green outlined arrow and the yellow outlined arrow in Figure 3(a).) Then, the DNVC moves only by the resonant contribution (indicated by the green filled arrow), i.e., the absorbing force. The absorbing force is proportional to the absorption of the particle. Thus, by observing the photon energy dependence of the moving distance of a particle, the absorption spectrum of a single particle can be evaluated. The sensitivity for extracting resonance component by NV-centers depends on the competition between the diffusion distance determined by environmental fluctuation and the distance by the force from the resonant component, as explained in Section 4 (Kinetic analysis).

Figure 3(b) shows the photon energy spectrum of the transport distance of the DNVC obtained by Brownian simulation and the fitting curve by Lorentz function. We assume room temperature ($T=300$ K), and the other environmental conditions are the same as in the previous subsection. The vertical axis represents the transport distance for 5 s for each photon energy of light wave 1. The distances are averaged over 10 trials of the transport. The resultant values are plotted in Figure 3(b) as green dots. We can see the influence of the thermal fluctuation though this is reduced by averaging over 10 trials. In this simulation, we assume that the resonance center and the dephasing constant of the NV-centers are $\omega_0 = 2.320$ eV and $\gamma = 20$ meV, respectively. The dashed line in Figure 3(b) represents the fitting curve by the Lorentz function. Interestingly, the values of the resonance center and the dephasing constant obtained in this fitting are $\omega_0 = 2.320$ eV and $\gamma = 19.35$ meV, respectively, which are in

Figure 3: (a) Schematic of the method to achieve a balance between the forces on the diamond as mother matrix (background forces) by resonant and non-resonant lights. By using two non-resonant light waves, it is possible to estimate the contribution of force arising solely from the diamond (outlined arrows) when the DNVC is irradiated by the resonant light wave. By using counter-propagating light waves that realize a balance between the background forces, we can extract the pure contribution of the resonance (green filled arrows.) (b) Photon energy spectrum of the transport

distance of DNVC obtained by Brownian simulation and the fitting curve by Lorentz function. The resonance center $\omega_0 = 2.320$ eV and the dephasing constant of the NV-center $\gamma = 19.35$ meV are obtained as fitting parameters. The original values were assumed as $\omega_0 = 2.320$ eV and $\gamma = 20$ meV.

good agreement with the original values we assumed for the model of NV-center in the simulation. This result indicates the possibility of obtaining the physical parameters such as absorption spectrum of single nanoparticles in good accuracy even in an aqueous environment, by observing their motions caused by the optical force owing to the resonant optical response. Note that not only the spectra but also the absolute values of absorption of individual nanoparticles can be obtained in principle by this method.

In actual experiment, the same information can be obtained by changing the intensity of light wave 1 to maintain the force balance of the NP, when the frequency is swept for light wave 1. The deviation between the intensities for the balance only for the background contribution and that including the resonance contribution provides information on absorption spectrum. The sensitivity of observation of the particle position and the extent of Brownian motion in the experiment determine the way that provides more accurate information.

2.3 Selective manipulation of QDs in a cryogenic condition

As reported in Ref. [23], liquid helium environment is one of the ideal conditions for optical manipulation because of its ultra-low viscosity. Furthermore, the resonance lines of the NPs become extremely sharp. Assuming the same condition as in Ref. [23], we theoretically demonstrate the selective transportation of QDs with narrow homogeneous line width by numerical simulation.

As illustrated in Figure 4(a), we consider the transportation of a particular size group of QDs with narrow

homogeneous absorption line width, with cancellation of the contributions from higher energy transitions of the other size groups of QDs using counter-propagating light waves. To demonstrate the essence of the proposed scheme, we consider simple two-level systems with different transition energies and same dipole moment. The absorption spectrum of each QD is assumed to consist of a single Lorentzian and the background. The latter is introduced by the quadratic function to represent the effect of higher energy transitions. For the material parameters, by considering those of CdSe QDs, we assume 36 Debye of dipole moment [44], and $\gamma = 30$ μ eV. We consider the assembly comprising 100,000 QDs, whose resonant energy is distributed in the range 1.94–2.00 eV (Group 1) and 2.52–2.58 eV (Group 2) based on a normal distribution, and the interval of resonant energy is 20 μ eV (Figure 4(b, c)). The diameter of QDs are assumed to be 3.0 nm for Group 1 and 1.75 nm for Group 2. In this scenario, the absorption edge of Group 2 is masked by the absorption spectra of Group 1, whose absorption edge lies in a significantly lower frequency region. Thus, selective manipulation of Group 2 using the usual optical manipulation method is difficult. Here, we show that highly sensitive selective manipulation of the masked QDs is possible by using counter-propagating light waves. For the present simulation, we assume that one incident light energy is 2.55 eV, which coincides with the transition energy of targeted QDs, and the other is 2.55 eV + 20 μ eV, which is on a slightly higher energy side. The light intensity is 1 kW/cm² for both light waves (Figure 4(a)).

Under the above conditions, the photon energy dependence of the optical force exerted on the particles is illustrated in Figure 4(d). This figure shows that a highly limited group of particles with a transition energy of

represented by dotted lines for reference.

approximately 2.55 eV receive a strong force. This is because the energy dispersion of absorption is rapid around the absorption edge, which enhances the difference between the forces induced by light wave 1 and 2. If the slope in the force spectrum of a QD between the photon energies of two light waves is negative (positive), a strong force directed toward positive (negative) z is induced. However, the dispersion in the other region is gradual, where the corresponding force difference becomes small, and the force direction is always toward negative z . Therefore, specific particles can be selected even in a scenario in which the spectral peak of the targeted particles is masked by the spectra of the other group of QDs with a lower energy absorption edge.

Under the above parameters, simulations were performed to see the transporting particles for 0.16 ms. We suppose the imaginary screen at the point $z = +7.0 \mu\text{m}$ (Figure 4(a)), to which the QDs adhere on arrival. The necessary transportation distance for isolating the targeted QDs can be estimated from the relationship between the

Figure 4: (a) Model of counter-propagating waves in liquid helium. We assume an imaginary screen at $z = +7.5 \mu\text{m}$ that can catch and fix the QDs. (b) Histograms of the number of QDs of the considered model. Total number of QDs for each peak structure is 100 000, and the energy interval is $20 \mu\text{eV}$. We assumed that the difference between resonance energies represent the difference in size and surface state of these two groups, and that the distribution of the number follows a normal distribution. (c) Inhomogeneous absorption spectrum of QDs. Red and green lines represent Group 1 and 2, respectively. They exhibit a set of QDs with slightly different resonance levels in each group, as seen in (b). The homogeneous width is assumed to be $30 \mu\text{eV}$. (d) Optical force along z -axis vs. transition energy of QDs, when we irradiate them with counter-propagating light waves with the photon energies of 2.55 and 2.55002 eV. (e) Absorption spectra of QDs that reached the screen after 0.16 ms irradiation of light waves. The peak structures indicate that the targeted QDs within the highly limited range of transition energy were selectively transported. The other small amounts of QDs around the sharp peak are considered to have reached the screen stochastically by Brownian motion. The spectra of all QDs are

transport distance by the optical force and the diffusion distance of QDs. This relationship is discussed in detail in Section 4 (Kinetic analysis).

Figure 4(e) shows the absorption spectrum of the particles reaching the screen. This result indicates that the particle assembly with a highly limited region of transition energy resonant with the incident light wave is selectively transported from the whole ensemble of particles. In this process, the QDs on which the strong negative force (appearing in the left half of the inset in Figure 4(d)) is exerted never reach the screen. Around the sharp peak, the absorption of a small amount of QDs can be seen. These small portions have reached the screen stochastically by Brownian motion. However, the small contribution in the higher energy side is attributed to the targeted QDs. To clarify the relationship with the whole assembly, the spectra of all particles are shown by dashed lines. In this result, we see that the QDs with absorption edge with lower energy side (Group 1) never reach the screen. Moreover, the counter-propagating waves can selectively manipulate a limited

group of QDs even if their resonance peaks are masked by the QD assembly with different absorption edges.

3 Conclusion

Optical manipulation using electronic resonance levels of NPs not only enhances the optical force but also realizes selective manipulation based on the quantum properties of NPs, which links the optical force technologies with materials science. However, manipulation using a single light wave by utilizing the resonance effect is difficult in actuality, because the resonance lines in the force spectra are hidden owing to various conditions. This problem can be addressed using multiple light waves, which cancel the forces arising from factors other than the resonance effect. This study theoretically demonstrated this concept for several typical cases in which selective resonance manipulation is difficult. The proposed method will significantly broaden the range of applications of resonant optical manipulation. Concentrating the particles possessing the desired quantum properties in a highly dilute solution is one of the important applications of the proposed method. Further, we should note that a method to extract the force component from solely the resonance contribution specific to the targeted quantum states can realize a new type of measurement of the single-particle optical response. The absorbing force on the matter system is generally proportional to the absorption of the system. Thus, we can obtain the direct information of matter absorption by observing the motion of single particles.

To clearly demonstrate the basic principle of the proposed concept, we used simplified models of targeted substance systems and the laser configuration, avoiding non-essential details. In the future, we will use a more sophisticated model for the simulation that enables more highly quantitative discussions. For experimental realization, some carefully devised environments are desired. For example, limiting the degrees of freedom of the particle motion using nano-channels or nano-fibers to reduce the fluctuations of particles and maintain sufficient light intensity over a long distance would be effective. Such experimental demonstrations performed by our group will be reported elsewhere.

4 Methods

4.1 Optical force

To calculate the optical force, we use the following general expression of the time-averaged optical force [19]:

$$\langle \mathbf{F} \rangle = (1/2) \text{Re} \left[\int_V d\mathbf{r} (\nabla \mathbf{E}(\mathbf{r}, \omega)^* \cdot \mathbf{P}(\mathbf{r}, \omega)) \right], \quad (1)$$

where $\mathbf{E}$ and $\mathbf{P}$ are the time-harmonic response electric field and induced polarization, respectively. Integration is performed over the volume of substances. We calculate the induced polarization $\mathbf{P}$ by solving the density matrix equation for the matter system including the phenomenological relaxation constants, as follows:

$$\hbar \frac{\partial \rho}{\partial t} = -i[H, \rho] - \Gamma \rho, \quad (2)$$

where ρ , H , and Γ denote the density matrix of the system, total Hamiltonian, and phenomenological relaxation constants, respectively. The Hamiltonian H is represented as $H = H_0 - \boldsymbol{\mu} \cdot \mathbf{E}(\mathbf{r}, t)$, where $\boldsymbol{\mu}$ denotes the operator of the dipole moment of the NP. Here, the first term H_0 denotes the unperturbed component (NP) and the second represents the light-NP interaction. We assume that the diagonal elements of $\boldsymbol{\mu}$ are zero. Then, we consider two time-harmonic plane waves, i.e., $\mathbf{E}_0(\mathbf{r}, t) = \mathbf{E}_1(\mathbf{r}, \omega_1) \exp(-i\omega_1 t) + \mathbf{E}_2(\mathbf{r}, \omega_2) \exp(-i\omega_2 t) + \text{c.c.}$, as the incident fields. Here, $(\mathbf{E}_1, \omega_1)$ and $(\mathbf{E}_2, \omega_2)$ represent the amplitude and the frequency of the first and second waves, respectively. In the present work, we consider up to the linear response; thus, the optical response occurs independently for each mode. The resonant part of the polarization in the linear response regime can be obtained as follows:

$$\mathbf{P}(\omega) = \sum_l |\boldsymbol{\mu}_l|^2 \left(\frac{1}{\omega_l - \omega - i\gamma_l} + \frac{1}{\omega_l + \omega + i\gamma_l} \right) \mathbf{E}(\omega), \quad (3)$$

where $\boldsymbol{\mu}_l$, ω_l , and γ_l are the dipole moment, electric resonant energy of NP, and damping for the transition between the l th state and the ground state, respectively, and ω is the frequency of the incident light.

Regarding the background polarization $\mathbf{P}_b$, we use the Clausius-Mosotti equation including the radiative reaction effect [45, 46], given by,

$$\begin{aligned} \mathbf{P}_b &= a_{\text{CMRR}} \mathbf{E}(\mathbf{r}, \omega), \\ a_{\text{CMRR}} &= \frac{a_{\text{CM}}}{1 - i q^3 \frac{a_{\text{CM}}}{6\pi\epsilon_0\epsilon_2}}, \\ a_{\text{CM}} &= 4\pi a^3 \epsilon_0 \epsilon_2 \frac{\bar{m} - 1}{\bar{m} + 2}, \end{aligned} \quad (4)$$

where ϵ_0 is the permittivity of vacuum, $\bar{m} = \epsilon_1/\epsilon_2$ (ϵ_1 and ϵ_2 are the dielectric constants of the target substance and surroundings, respectively), q is the wavenumber of the incident light wave, and a is the radius of the target particle.

By substituting the sum of $\mathbf{P}(\omega)$ in (3) and $\mathbf{P}_b$ into (1) as the whole polarization along with $\mathbf{E}_0$, we obtain the optical force $\langle \mathbf{F} \rangle$ exerted on an NP. (In the present demonstration, we use an incident field $\mathbf{E}_0$ for $\mathbf{E}$ because the effect of the scattered field is negligible in the considered particle size regime.)

For a single component light wave, we define $\mathbf{E}_i$ as,

$$\begin{aligned} \mathbf{E}_i(\mathbf{r}, \omega) &= \mathbf{E}_{i0} \exp(i(\mathbf{q} \cdot \mathbf{r})) \\ |\mathbf{E}_{i0}|^2 &= \frac{2I}{c\epsilon_2\epsilon_0} \end{aligned} \quad (5)$$

where $i = 1, 2$, I is the intensity of the incident light wave, and c is the speed of light in vacuum.

In addition, the evanescent wave at the solid-liquid interface is given by,

$$\mathbf{E}_t = \mathbf{A}_t \exp \left[-\frac{\omega}{c} \sqrt{n_s^2 \sin^2 \theta_i - n_l^2} x + i \left(\frac{\omega}{c} n_s \sin \theta_i z - \omega t \right) \right], \quad (6)$$

where $\mathbf{A}_t$ is the transmission electric field coefficient, n_s and n_l are the solid and liquid refractive indices, respectively, and θ_i is the incident

angle. We used this equation to treat the evanescent wave for the incident light.

4.2 Kinetic analysis

To investigate the feasibility of selective transportation using counter-propagating light waves, we perform kinetic analysis with Brownian motion in a solvent using the Langevin equation, as follows [47]:

$$m \frac{d^2 \mathbf{r}(t)}{dt^2} = \mathbf{F} - \gamma \frac{d\mathbf{r}(t)}{dt} + \mathbf{F}_{\text{random}}, \quad (7)$$

where, $\mathbf{r}$ is the position of NP of mass m , $\mathbf{F}$ represents the optical force, $\gamma = 6\pi\eta a$ (η : viscosity of media, a : radius of particle), and $\mathbf{F}_{\text{random}}$ is a random force. For water as the surrounding medium ($T=300$ K), we use $\eta = 0.89 \times 10^{-3}$ Pa·s, and for liquid helium ($T=2$ K), we use $\eta = 1.468 \times 10^{-6}$ Pa s as the experimental values [48].

If the time step size Δt is sufficiently longer than the momentum relaxation time ($\Delta t \gg m/\gamma$), (7) can be rewritten as,

$$\mathbf{r}(t + \Delta t) = \mathbf{r}(t) + \frac{\Delta t}{\gamma} \mathbf{F} + \Delta \mathbf{r}_{\text{ran}}, \quad (8)$$

here, the effect of thermal fluctuation is rewritten into the random displacement $\Delta \mathbf{r}_{\text{ran}} = (\Delta x_{\text{ran}}, \Delta y_{\text{ran}}, \Delta z_{\text{ran}})$, which satisfies the following equation:

$$\langle \Delta x_{\text{ran}} \rangle = \langle \Delta y_{\text{ran}} \rangle = \langle \Delta z_{\text{ran}} \rangle = 0, \quad (9)$$

$$\langle (\Delta x_{\text{ran}})^2 \rangle = \langle (\Delta y_{\text{ran}})^2 \rangle = \langle (\Delta z_{\text{ran}})^2 \rangle = \frac{2k_B T}{\gamma} \Delta t. \quad (10)$$

In the above expression, k_B is the Boltzmann constant, and T is the temperature of the medium. The random displacement $\Delta \mathbf{r}_{\text{ran}}$ follows normal distribution.

Using the Box-Muller method as a random number generation method, the random displacement $\Delta \mathbf{r}_{\text{ran}}$ is given by,

$$\Delta \mathbf{r}_{\text{ran}} = \sqrt{-2 \langle (\Delta \mathbf{r}_{\text{ran}})^2 \rangle \log X} \cos(2\pi Y) \Delta t, \quad (11)$$

where X, Y is a random variable (random number) consisting of uniform distribution within (0, 1).

By using optical force as an external force, the manipulation of particles in the medium can be calculated. Here, we ignore the vortices and flow in the fluid, and express the Brownian motion based on the dynamic viscosity and temperature in the fluid. Therefore, it is possible to calculate for the medium which we can obtain viscosity coefficient experimentally.

Assuming that a single particle in the solvent receives an external force and reaches the terminal velocity in a very short period, the transport distance X_h for a certain time and the diffusion distance X_d for the same time by the Brownian motion are described as follows:

$$X_h = \frac{F}{6\pi\eta a} t, \quad (12)$$

$$X_d = \sqrt{\frac{k_B T t}{3\pi\eta a}}. \quad (13)$$

Figure 5 expresses this equation graphically, where the spatial range of diffusion of particles by the Brownian motion is represented by concentric circles expanding with time. On the other hand, because of the transportation by the external force, the center of the circle representing this reachable range shifts in the direction of the external force. If these reachable areas overlap, it cannot be distinguished whether the particles were transported by the external force or accidentally moved to that place by Brownian motion. Therefore, to perform sorting by transport, the transport distance by the external force must exceed the range of Brownian motion; furthermore, we obtain

$$X_h - X_d > X_d \Leftrightarrow \frac{X_h - X_d}{X_d} > 1 \Leftrightarrow \frac{X_h}{X_d} > 2 \quad (14)$$

as a conditional expression.

Figure 5: Conceptual diagram of movable range of particles. Yellow concentric circles and blue circle represent the areas where particles are moved by diffusion and by diffusion and external force, respectively. The center of the circle shifts in the direction of the external force. If particles exist in the area where the yellow and blue circles overlap, it cannot be distinguished whether the particles were carried by the external force or were accidentally moved there by Brownian motion. Therefore, to perform sorting by transport, the transport distance by external force must exceed the range of the Brownian motion. From this condition, it is possible to obtain an approximate particle travel distance for the time required for transport.

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