Proposed scheme of realization of artificial nano-heterostructures by optical force under double resonance

Yuto Yamada\textsuperscript{1}, Masayuki Hoshina\textsuperscript{2}, Tomohiro Yokoyama\textsuperscript{1}, and Hajime Ishihara\textsuperscript{1,2}

\textsuperscript{1} Division of Frontier Materials Science, Osaka University, 1-3 Machikaneyama-cho, Toyonaka, Osaka 560-8531, Japan
\textsuperscript{2} Department of Physics and Electronics, Osaka Prefecture University, 1-1 Gakuen-cho, Naka-ku, Sakai, Osaka 599-8531, Japan
E-mail: yamada.y@opt.mp.es.osaka-u.ac.jp

Abstract. We theoretically study a new configuration control of nanoparticles based on an optical force, which enables to trap two kinds of nanoparticles simultaneously in two-dimensional hetero-arrangement. Our proposed scheme utilizes two resonance effects on a localized surface plasmon and an electronic transition of nanoparticles. These resonances change the optical force behavior significantly. We propose periodic rhomboid metallic nanostructures with two linear polarized laser beams, the directions of which polarization are parallel to the two diagonals of rhomboid. The polarization direction fractionates the trapped nanoparticles when the two laser frequencies correspond to the electronic transition energies of two kinds of particles, respectively. We evaluate the feasibility of our proposed scheme numerically. The map of the optical force shows a clear spatial separation of trapping position for the two kinds of particles. Our results demonstrate an essential advantage of the optical manipulation that enables creating novel nanostructures unrealizable by conventional nanofabrication techniques.

1. Introduction
Optical manipulation is a technique to manipulate minute objects by using an optical force. It has been applied for micrometer-scale objects since A. Ashkin and his collaborators have demonstrated the trapping of particles with a single laser beam in 1986 [1]. In recent years, attempts to manipulate nanometer-scale objects by the optical force have been advanced. Nanoparticles smaller than the wavelength of visible light exhibit significant quantum effects which are hardly observed in micrometer-size objects. Thus, a development of control technology of nanoparticle’s motion and position within nanometer-scale resolution will open a new perspective to obtain unprecedented, indented, and useful functions by configuration and ordered structures of them. From this point of view, the optical manipulation has advantages for the operation over a large-area in a non-contact manner. However, the optical manipulation of nanoparticles is still a challenging task because of a weak exerted force, which is approximately proportional to the volume of the target within the Rayleigh scattering regime. As strategies for the nanoscale optical manipulation, the localized surface plasmon (LSP) resonance and the electronic resonance of nanoparticles are key ingredients [2,3]. The LSP is a collective oscillation of electrons on the surface of metallic structures. Its resonant frequency is sensitive to the size and shape of metallic structures. The LSP for a nanoscale metallic-gap results in an extremely
enhanced electric field at the gap (plasmonic hotspot). This strong and localized electric field attracts nanoparticles toward the hotspot strongly and achieves a nanometer-scale resolution. The optical force is also enlarged when the frequency of applied field is resonant with the electric transition of nanoparticles. The resonance has also an advantage for a selective manipulation of nanoparticles with quantized electronic levels.

In this work, we theoretically demonstrate a new function of optical manipulation based on an interplay of two resonance effects for the LSP and the electronic transition of nanoparticles. The schematic view of our proposed system is illustrated in Fig. 1. We consider a periodically arranged rhomboid metallic nanostructure and two linear polarized laser beams, labeled as A and B. We suppose that the incident fields are plane waves. The direction of polarization of A is parallel to one diagonal of rhomboid metal. That of B is along to the other diagonal. We investigate a simultaneous optical trapping of two kinds of nanoparticles by this system. The polarization direction of two beams selects trapping positions for nanoparticles. Then, the optical force distributes nanoparticle a and b as shown in Fig. 1. The LSPs induced by light A and B show different resonant frequencies due to the rhomboidal shape of metal. To see a clear distribution, we assume that the electronic transition energies of nanoparticle a and b are close to the resonant energies of LSP by light A and B, respectively. Such assumption can be satisfied by proper fabrication of metallic nanostructure. We demonstrate a numerical calculation based on the discrete dipole approximation (DDA) method [6] and discuss an absorption cross section of the metals and the optical force on the particles to evaluate the feasibility of our proposal.

2. Theoretical model

In the numerical demonstration, we evaluate a time-averaged optical force \( \langle F \rangle \) [4] given by a total electric field \( \mathbf{E}(r,t) = \mathbf{E}(r)e^{-i\omega t} \),

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

where the integration is taken for the whole of nanoparticle. \( \mathbf{P} \) is the induced polarization. In addition to \( \langle F \rangle \), we obtain the absorption cross section \( \sigma_{\text{abs}} \) from \( \mathbf{E} \) as \( \sigma_{\text{abs}} = 4\pi k \int_V \mathrm{d}r \frac{\lvert \mathbf{E}(r)/\mathbf{E}_0(r) \rvert^2 \text{Im}[\chi(r; \omega)]}{\lvert \mathbf{E}(r)/\mathbf{E}_0(r) \rvert^2 \text{Im}[\chi(r; \omega)]} \), where \( k \) is the wave number of incident field [5]. The total electric field is obtained from the Maxwell’s equation and a constitutive equation. The latter is \( \mathbf{P}(r) = \chi(r; \omega)\mathbf{E}(r) \) with a local susceptibility \( \chi(r; \omega) \). A solution of the Maxwell’s equation is given by a free-space Green’s function \( \mathbf{G}(r, r'; \omega) \) as \( \mathbf{E}(r) = \mathbf{E}_0(r) + \int \mathrm{d}r' \mathbf{G}(r, r'; \omega)\mathbf{P}(r') \), where \( \mathbf{E}_0 \) is an incident electric field. Substitution of the constitutive one into this Maxwell’s equation provides a self-consistent equation that determines the total electric field \( \mathbf{E}(r) \).

For complicated structures, we applied the DDA method [6] to solve the self-consistent equation. The DDA discretizes a 3D space to cubic cells. Then, the self-consistent equation becomes a set of simultaneous linear equations for \( \mathbf{E} \). The resolution of numerical calculation is restricted by the size of cells. Let us put the cell size as 1 nm. For the DDA calculation, we apply the Drude model to the susceptibility of metal with the parameters of gold. The
nanoparticles are described by the Lorentz model using the parameters of typical fluorescent dye, where the resonance energies are 1.35 eV and 1.75 eV for a and b, respectively, and the damping constant is 1 meV. We apply a point dipole approximation to the nanoparticles with 9.7 D expressed by a single cell in the DDA. The parameters are summarized in Fig. 2. The incident fields are plane waves. As a medium, we assumed a vacuum.

3. Results and discussions
We perform the calculation when only four metallic nanostructures are present (see Fig. 2). Note that the length of all four gaps between the metals are equivalent with each other.

First, we investigate the absorption cross section of four metals in the absence of nanoparticles. Figure 3a shows the spectrum of $\sigma_{abs}$ of four metals when either laser A or B is irradiated. The spectrum depends clearly on the direction of incident polarization. The difference of resonant energy is attributed to the difference of diagonal of rhomboid metal. This anisotropy agrees qualitatively with the LSP on prolate spheroidal metals [7]. Figure 3b shows the profiles of intensity of total electric field for an incident of light A at 1.35 eV and that of light B at 1.75 eV. Light A (B) excites long (short) diagonal’s nanogaps with different frequencies, and does not induce strong electric field at the others.

Figure 4 exhibits a map of optical force exerted on a single particle a (red) or b (blue arrow) when both light A and B are irradiated simultaneously. Here, the energies of incident light A and B are slightly smaller than the resonace of particle a and b, respectively (light A: 1.3484 eV, B: 1.7484 eV) to obtain stronger gradient force reflecting a real part of Lorentz function. The intensity of incident electric fields is 1 kW/cm$^2$. We find that the optical force on particle a is strong in the vicinities of nano-gaps for long diagonals, whereas the force on b is for short diagonals. The difference of magnitudes is due to the difference of absorption cross section because of the shape anisotropy of metals. The clear separation of the forces distribution enables selective and simultaneous arrangement of two kinds of nanoparticles.

4. Conclusions
We have studied a simultaneous and selective optical trapping for two kinds of nanoparticles based on our proposed metallic nanostructure. There are three key ingredients: rhomboidal shape of metals, irradiation of two linear polarized lights in different directions, and interplay of the resonances on the LSP and nanoparticles. We have demonstrated a numerical simulation based on the DDA method and exhibited enough feasibility for the simultaneous trapping of two kind of particles to realize hetero-arrangement of them.

Our proposed structure would be extended for more than two kinds of nanoparticles and more complicated arrangement. Such variety of nanoparticle’s arrangement leads to novel and high-performance functions by applying to a material surface.
Figure 3. (a) Absorption cross section of the system comprising rhomboid metals shown in Fig. 2 when light A (red line) and light B (blue) are irradiated. The dashed lines indicate the energy 1.35 eV and 1.75 eV. (b) Profiles of intensity of the total electric field at 1.35 eV for light A (left panel) and at 1.75 eV for light B (right). The other parameters are mentioned in Fig. 2.

Figure 4. Map of the optical force exerted on a single nanoparticle under the irradiation both of light A and B. Red and blue arrows correspond to the particle a with resonance at 1.35 eV and the particle b at 1.75 eV, respectively. The length of arrows means the magnitude of the force. The gray dashed line indicates the edge of metallic nanostructures. The other parameters are mentioned in Fig. 2. The force weaker than 0.2 fN is not shown.

Acknowledgments
We acknowledge supports from JSPS KAKENHI Grant No. JP16H06504 in Scientific Research on Innovative Areas: “Nano-Material Optical-Manipulation”.

Reference
[1] Ashkin A, Dziedzic J M, Bjorkholm J E, and Steven Chu 1986 Opt. Lett. 11 288-290.
[2] Tsuboi Y, Shoji T, Kitamura T, Takase M, Murakoshi K, Mizumoto Y, and Ishihara H 2010 J. Chem. Lett. 1 2327.
[3] Iida T and Ishihara H 2003 Phys. Rev. Lett. 90 057403.
[4] Iida T and Ishihara H 2008 Phys. Rev. B 77 245319.
[5] Goedecke G H and O’Brien S G 1988 Appl. Opt. 27 2431.
[6] Purcell E M and Pennypacker C R 1973 Astrophys. J. 186 705.
[7] Barber P W, Chang R K, and Massoudi H 1983 Phys. Rev. Lett. 50 997.