Trapping and Deposition of Dye–Molecule Nanoparticles in the Nanogap of a Plasmonic Antenna

Christophe Pin,† Shutaro Ishida,† Genta Takahashi,† Kota Sudo,† Tuyoshi Fukaminato,‡ and Keiji Sasaki‡,*

†Research Institute for Electronic Science, Hokkaido University, Kita 20 Nishi 10, Kita-ku, Sapporo 001-0020, Japan
‡Department of Applied Chemistry & Biochemistry, Graduate School of Science & Technology, Kumamoto University, 2-39-1 Kurokami, Chuo-ku, Kumamoto 860-8555, Japan

ABSTRACT: Plasmonic nanostructures, which allow light focusing at the deep subwavelength scale, and colloidal nanoparticles with unique optoelectronic properties are nowadays fabricated with nanometer precision. However, to fully control and exploit nanoscale light–matter interactions in hybrid plasmonic–nanophotonic devices, both materials must be assembled in heterostructures with similar precision. Near-field optical forces have recently attracted much attention, as they can precisely trap and position nanoparticles at plasmonic hotspots. However, long-range attraction and the surface bonding of nanoparticles usually require other specific techniques, such as electrothermal heating and surface chemical treatments. This Letter reports on the optical trapping and deposition of dye–molecule nanoparticles in the nanogap of a gold antenna. The nanoparticles are captured by focusing a near-infrared laser beam on a targeted plasmonic antenna. This single-step deposition process requires only a few seconds under 1.4–1.8 MW·cm⁻² continuous-wave illumination and shows a polarization dependence smaller than expected. Fluorescence and electronic microscopy observations suggest that nanoparticle deposition arises from a trade-off between optical and thermal effects.

INTRODUCTION

The possibility of assembling metallic and dielectric materials into nanoscale heterostructures has recently attracted much attention.¹ Specific materials integrated into metallic nanostructures such as plasmonic antennas and waveguides can enhance nanoscale light–matter interactions such as directional light coupling and emission,²−⁴ surface-enhanced Raman scattering (SERS),⁵ and infrared absorption (SEIRA),⁶ enhanced and tunable single- and two-photon absorption/emission,⁷−¹² second- or third-harmonic generation (SHG, THG),¹³,¹⁴ and strong plasmon–exciton coupling.¹⁵ These near-field phenomena have given birth to numerous new fabrication and characterization techniques and to a new class of hybrid plasmonic–nanophotonic components.¹ Among the candidate integration materials, organic molecules are particularly anticipated in miniaturized devices with improved or new functionalities for molecular nanoelectronics and nanophotonics applications.¹,¹⁸−²⁰

However, fabricating metal–dielectric nanostructures, especially those involving plasmonic nanogap features, remains a challenging task. Whereas downscaled top-down processes inherited from microelectronics face critical resolution limits, bottom-up approaches promise to simplify and reduce the cost of nanofabrication techniques, while preserving their high accuracy, reliability, and reproducibility. A promising avenue is the direct assembly of liquid-suspended nanomaterials on targeted points on a chip’s surface.¹¹,¹³,¹⁶,²¹−²⁶ Colloidal synthesis science has been actively and fruitfully researched in recent years, enabling the fabrication of miscellaneous kinds of nanoparticles, from metallic and carbon nanostructures to quantum dots and molecular aggregates. In this technique, the compositions, sizes, and shapes of the particles can be tightly controlled.

Several deposition techniques have recently flourished. Among these, deposition methods based on optical forces can precisely attach a large variety of nanomaterials to target surfaces.²⁷−³⁵ Especially, near-field optical gradient forces arising from localized surface plasmons can be conveniently used to attract nanoparticles toward plasmonic hotspots¹⁶−³⁹ and precisely fix the particles at the trap location.²³,²⁶ Nevertheless, the deposition requires accurate nanoscale management of optical, electrostatic, chemical, and thermal interactions. For instance, particle adhesion and long-range attraction have been promoted in previous works by chemical-surface functionalization and electrically induced heating, respectively.²⁰,²³

In this Letter, we introduce a fast single-step technique for the bottom-up fabrication of hybrid plasmonic–nanophotonic...
devices. More specifically, we show that organic dye–molecule nanoparticles suspended in a liquid colloidal solution can be precisely deposited in the nanogap of a plasmonic bowtie antenna under intense laser irradiation.

**RESULTS**

The deposition technique is demonstrated on nanoparticles of orange organic dye molecules (1,4-bis(4-diphenylaminophenyl)-2,1,3-benzothiadiazole) fabricated by the reprecipitation method.\(^{40,41}\) The 2,1,3-benzothiadiazole (BTD) fluorophore exhibits strong fluorescence properties in both solution and solid (nanoparticle) states, a large Stokes shift, and high photo-stability.\(^{42}\) It also forms stable spherical nanoparticles in aqueous solution.\(^{43}\) In the present study, the BTD molecules were dissolved in acetone (1 \(\times\) \(10^{-3}\) mol \(L^{-1}\)), and 0.3 mL of this solution was quickly dropped into 9 mL of ultrapure water (Millipore Direct Q UV3 Water Purification System) with vigorous stirring, as shown in Figure 1a. After 10 min of continuous stirring, the residual organic solvent (acetone) was removed from the obtained solution by heating in a microwave oven (2.45 GHz, 500 W, 2 min).

Figure 1b shows a scanning electron microscopy (SEM) image of the nanoparticles contained in a drop of the solution that was deposited and dried on a glass coverslip. To prevent charging effects, a few-nanometer-thick layer of tungsten was deposited on the sample. Most of the observed single nanoparticles are 30–40 nm wide (average diameter = 34 nm). The minimum and maximum values of measured particle diameters were 22 and 57 nm, respectively (see Supporting Information). However, the number of small particles may have been underestimated because of the limited SEM resolution and the high density of the nanoparticles on the SEM images.

The plasmonic nanogap antennas in this work were gold triangular dimers, called bowtie nanoantennas. These nanoantennas were fabricated on a glass substrate (Matsunami #5) by electron beam (e-beam) lithography. After e-beam exposure and development of the resist film, a 30 nm thick gold layer was deposited on a 3 nm thick chromium adhesion layer. After lift-off, bowtie antennas with nanogaps of only a few nanometers were successfully fabricated. Each gold nanotriangle had a 250 ± 5 nm long edge orthogonal to the antenna axis and a 230 ± 5 nm length along the antenna axis.

A thin microfluidic chamber was built on a thin glass coverslip (Matsunami #1) as follows. The colloidal nanoparticle solution was sonicated, and a 5 \(\mu\)L drop of the sonicated solution was deposited on the thin coverslip. The coverslip was then covered with the plasmonic chip in an inverted configuration (with the plasmonic structures located on the bottom side). Finally, the fluidic chamber was sealed with tape to prevent water evaporation. Based on the dimensions of the plasmonic chip (24 × 24 mm\(^2\)), the estimated depth of the water chamber was 8 \(\mu\)m.

The sample was then placed on an inverted microscope stage (see Figure 2) and targeted with near-infrared (NIR) light from a Nd:YAG laser source (Millennia IR, Spectra-Physics) emitting at 1064 nm. The light was focused through a high-numerical aperture lens (Olympus UPlanSApo 100 × /1.40 oil). The fluorescence of the nanoparticles was excited by a green laser emitting at 532 nm at low power (laser intensity below 0.1 kW cm\(^{-2}\)). To prevent rapid photobleaching of the dye molecules, the fluorescence excitation was modulated by a signal generator. The incident laser beams are directed toward the sample by two dichroic mirrors. The fluorescence emission is passed through a bandpass filter and collected by an intensified charge-coupled device (ICCD) camera.
focal point of the NIR laser, then returning it to its initial position. This process was completed within a brief time (less than 5 s). When the nanoantenna reached the focal point of the laser beam, the laser intensity jumped from an estimated 55−74 kW cm$^{-2}$ to 1.4−1.8 MW cm$^{-2}$. During this brief increase in laser irradiation intensity, a few of the nanoparticles were attracted and deposited in the nanogap of the targeted antenna. Particle adhesion on the sample surface required a strong laser intensity; at lower intensities, the optical trapping of nanoparticles was brief and reversible.

Figure 3b shows an image sequence from Movie S1, illustrating the nanoparticle-deposition process described above. Initially, the imaging plane coincided with the sample surface, and the defocused NIR laser beam was aligned with the bare gold nanoantenna. Only the fluorescent light emitted by previously deposited nanoparticles at the right side of the picture can be observed. Eventually, the sample surface moves through the focal point of the NIR laser beam. The fluorescent signal from the previously deposited nanoparticles decreased as the imaging plane shifted downward, and a weak fluorescent signal appeared at the laser spot position. Within 5 s, the focusing condition was restored to its initial stage. Nanoparticle deposition is evidenced by moving the gold antenna away from the laser beam path. Scale bar: 2 μm.

Figure 4. SEM micrographs of deposited nanoparticles (false-color representation). (a−c) Nanoparticles deposited under parallel polarization at different irradiation intensities. The nanoparticles are successfully deposited in the nanogaps of the gold antennas at 1.4 MW cm$^{-2}$ and 1.6 MW cm$^{-2}$ but are melted at each side of the nanoantennas at 1.8 MW cm$^{-2}$. (d) A bare gold nanoantenna, shown as a reference. Scale bar: 100 nm. (e) Nanoantenna without deposited nanoparticle after irradiation under transverse polarization conditions at 1.6 MW cm$^{-2}$. (f) Single nanoparticle deposited in the nanogap of a gold antenna under transverse polarization conditions at 1.8 MW cm$^{-2}$.
elliptically polarized laser beam at three intensity settings. Because the incident beam was partially depolarized after reflection by the band-pass dichroic mirror, the polarization ratio in the nanoparticle-deposition experiments were set to 10:1. The polarization was set either parallel or orthogonal to the antenna’s main axis. Under the parallel polarization condition, a few nanoparticles were trapped and deposited in the nanogaps of the bowtie antennas at 1.4 and 1.6 MW cm$^{-2}$ laser intensities, as can be observed in panels a and b of Figure 4, respectively. However, at higher laser intensity (1.8 MW cm$^{-2}$), the nanoparticles attracted toward the antenna were stopped before reaching the nanogap. Some nanoparticles (shown in red in Figure 4c) were deposited and melted a few tens of nanometers away from the nanogap on both sides of the plasmonic antenna. On the other hand, under the transverse polarization condition, no particle was deposited at 1.4 and 1.6 MW cm$^{-2}$ (Figure 4d), but interestingly, single nanoparticle deposition was achieved at 1.8 MW cm$^{-2}$ (Figure 4f). This result proves that the optical gradient force was sufficiently strong to attract nanoparticles toward the antenna’s nanogap using high laser intensity irradiation. In other words, sufficient electromagnetic energy was coupled to the plasmonic gap mode, probably because the incident beam was elliptically polarized. No particle was found to be deposited at the outer corners of the gold nanostructure.

It can be seen in the panels a, b, and f of Figure 4 that the nanoparticles were partially melted when adhering to the nanogap surface. Despite the shape modification caused by the partial melting, the particles can be individually distinguished even when forming a compact assembly in the nanogap vicinity. The sizes of the deposited nanoparticles well agree with the nanoparticles’ size measured prior to the experiment (see Supporting Information). Although the nanoparticles are larger than the nanogap’s size, it can be seen that the nanogap was filled by some melted parts of the deposited nanoparticles during the deposition process. Thus, the partial melting of the deposited nanoparticles allows for the deposition of dye molecules in the nanogap of the plasmonic antenna.

### DISCUSSION

Although it was already reported in the literature that plasmonic nanoantennas can be used to trap nanoparticles,36–39 nanoparticle deposition relying on near-field optical gradient forces arising from surface plasmons remains a widely unexplored field. When a nanoparticle is trapped by a plasmonic nanoantenna, the particle can be adsorbed at the surface of the antenna if the optical gradient force acting on the particle is strong enough to overcome the electrostatic repulsion between the particle’s and sample’s surfaces. When the distance between both surfaces shrinks to a few nanometers, the two surfaces attract each other and permanently bond under attractive van der Waals interactions. In this work, the partial melting of the deposited nanoparticles leads to an extended contact area between the particle and the sample, which may play an important role in the adhesion of the nanoparticles.

The partial or complete melting of the deposited nanoparticles observed on the SEM images indicates that the temperature significantly rises near the surface of the gold nanonatenna during the deposition process. The heat generated by the NIR light absorbed by the gold nanostructure plays a significant role in the nanoparticle trapping mechanism, causing additional effects such as heat convection, thermophoresis, and thermo-osmosis. As discussed hereinafter, these phenomena may either help or hinder the optical trapping of the nanoparticles.

As previously mentioned, under the parallel polarization conditions the nanoparticles attracted toward the nanoantenna were stopped and deposited before reaching the plasmonic nanogap when the laser intensity was increased to 1.8 MW cm$^{-2}$ (Figure 4c). Tsuibo et al., who observed a similar phenomenon at the microscale,27 suggested that such deposition patterns result from negative thermophoretic forces acting on the nanoparticles. At high laser intensities, repulsive thermophoretic forces may counterbalance the attractive optical gradient force as the temperature of the gold antenna reaches a given threshold. Consequently, the nanoparticles cannot reach the nanogap of the antenna and are instead deposited at the position of equilibrium between both thermophoretic and optical gradient forces.

The heat diffusion in the liquid environment of the gold antenna may also generate hydrodynamic flows, via convection or thermo-osmosis. Although previous work from Ndukaife et al. reported that a controlled convective flow can advantageously be used for the long-range attraction of particles,26 convective flows are usually considered as undesirable phenomena, making optical trapping unstable or even impossible. In our experiment, the 8 μm high microfluidic chamber was designed for preventing convective flows from reaching more than a few tens of nm s$^{-1}$.44 Yet, the nanoparticles were trapped and deposited within seconds, in contrast to low-power plasmonic trapping of larger particles (which rather occurs on minute time scales). Given the low nanoparticle concentration (only a few nanoparticles were visible at any one time in the (41 × 28) μm$^2$ field of view) and the limited range of action of the near-field optical gradient force, thermal effects are expected to play a fundamental role in the fast, long-range attraction process, drawing the nanoparticles toward the plasmonic antenna over distances of several micrometers.

Heat convection being ruled out, the long-range attraction of the nanoparticles was attributed to thermo-osmosis, a phenomenon already reported by different groups.45,46 Thermo-osmotic flows result from the gradient of surface tension induced by a temperature gradient at the interface between two phases (the liquid water and the glass substrate in the present case). Due to the nanosize and the high temperature of the gold antenna, a thermo-osmotic flow may be generated during the deposition process, drawing surrounding particles toward the center of the gold nanoantenna. Considering the time scale of the nanoparticle trapping and deposition mechanism, the thermo-osmotic flow velocity is expected to reach at least a few tens of micrometers per second in our experiment, which is in good agreement with already observed thermo-osmotic flows.45,46 Such a fast flow converging toward the nanogap of the antenna may also explain the absence of particle deposited near the outer corners of the gold nanostructure. Indeed, the temperature gradient is expected to be stronger around the outer tips of the gold triangles, while the temperature gradient near the nanogap should tend to zero as the gap size decreases. Thus, the flow velocity near the outer corners of the antenna is expected to be faster than the flow velocity in the vicinity of the nanogap. Moreover, due to its central position below the circulating fluid loop, the nanogap should be subject to slower fluid flow.46 This would make the trapping of nanoparticles more unstable in the outer part of the
antenna, while still allowing nanoparticles to be trapped and
deposited in the nanogap of the antenna.

In addition, it is worth noticing that as recently highlighted
by Baffou et al., the amount of heat induced by the light
absorbed by the gold nanostructure is not directly proportional
to the electromagnetic field enhancement in the nanogap of the
antenna. This fact implies that the heat-induced long-range
attraction and the optical gradient force-induced short-range
attraction of the particles are not directly correlated. The NIR
laser light being absorbed under both polarization conditions,
the light polarization should have a weaker influence on the
long-range attraction of the nanoparticles than on the short-
range attraction. In the case of the long-range attraction being
the critical factor for the particle deposition to occur, it seems
consistent to observe the deposition of nanoparticles at
relatively close laser intensities under both polarization
conditions.

Eventually, we would like to underline the fact that despite
the small amount of deposited fluorescent material a bright
fluorescence signal was still experimentally observed after
the nanoparticles had settled in the nanogaps of the plasmonic
antennas. The dye molecules used in this work are robust
against fluorescent quenching effects, enabling the detection of
nanoparticle-deposition events to the single-particle level. As
the described technique requires no chemical surface
functionalization prior to the nanoparticle deposition, we believe
that it is straightforwardly adaptable to the deposition of
other organic nanomaterials. Nonorganic nanoparticles may be
deposited in a similar way, depending on their surface
properties. However, trapping and depositing smaller nano-
particles would require a substantial increase of the laser
intensity, which may result in detrimental heating effects.

**CONCLUSIONS**

In summary, we have experimentally demonstrated a one-step
optical process for the trapping and the deposition of dye–
molecule nanoparticles in the nanogaps of plasmonic nano-
antennas. This process operates by briefly increasing the
irradiation intensity of a laser focused on a gold bowtie
nanoantenna. Nanoparticles with approximate diameters of
30–40 nm were successfully attracted and deposited at the
centers of the gold antennas. Consequently, plasmonic gaps less
than 20 nm wide were filled with nanoscale amounts of organic
material in a single optical operation. While precise positioning
and adhesion of the nanoparticles relied on the action of the
near-field optical gradient force, the fast, long-range attraction
of the nanoparticles was attributed to light-absorption-induced
thermo-osmotic flows in the thin microfluidic chamber. This
technique allows the precise deposition of colloidal nano-
particles in targeted plasmonic nanogaps, bringing new
perspectives to the fabrication of on-chip hybrid plasmonic–
nanophotonic devices. For instance, the hybrid nanoparticle–
antenna structures fabricated in this work could be used as
efficient nanoscale organic light sources in integrated plasmonic
circuits. With additional specific functionalization of the
nanoparticles, this technique may also find applications for
the fabrication of a highly ordered array of organic nano-
particles for ultrahigh density optical memory devices, highly
sensitive sensors, and so on.

**ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge on the
ACS Publications website at DOI: 10.1021/acsomega.8b00282.

Fluorescence excitation, emission, and absorption spectra
of the fabricated dye–molecule nanoparticles, SEM micrographs with size measurements of the fabricated
nanoparticles and deposited nanoparticles, and original
version of the SEM micrographs shown in Figure 4.

Video of the nanoparticle-deposition process observed by
fluorescence microscopy. Scale bar: 5 μm (AVI)

**AUTHOR INFORMATION**

**Corresponding Author**

*E-mail: sasaki@es.hokudai.ac.jp. Tel.: +81-11-706-9396. Fax:
+81-11-706-9391.

**ORCID**

Christophe Pin: 0000-0002-5088-5711

**Author Contributions**

The manuscript was written through contributions of all
authors. All authors have given approval to the final version of
the manuscript.

**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

We acknowledge financial support from JSPS KAKENHI for
Scientific Research on Innovative Areas (Grant Number
JP16H06504, “Nano-Material Optical-Manipulation”). C.P.
acknowledges funding from the Research Institute for
Electronic Science, Hokkaido University.

**ABBREVIATIONS**

BTD, 1,4-bis(4-diphenylaminophenyl)-2,1,3-benzothiadiazole;
SEM, scanning electron microscopy; NIR, near-infrared; ICCD, intensified charge-coupled device

**REFERENCES**

(1) Benson, O. Assembly of hybrid photonic architectures from
nanophotonic constituents. *Nature* 2011, 480, 193–199.
(2) Curto, A. G.; Volpe, G.; Tamiaiu, T. H.; Kreuzer, M. P.;
Quidant, R.; van Hulst, N. F. Unidirectional Emission of a Quantum
Dot Coupled to a Nanotenna. *Science* 2010, 329, 930–933.
(3) Vercruyssse, D.; Zheng, X.; Sonnensfraud, Y.; Verellen, N.; Di
Martino, G.; Lagae, L.; Vandenbosch, G. A. E.; Moshchalkov, V. V.;
Maier, S. A.; Van Dorpe, P. Directional fluorescence emission by
individual V-antennas explained by mode expansion. *ACS Nano* 2014,
8, 8232–8241.
(4) Kim, J.; Song, J. H.; Jeong, K. Y.; Ee, H. S.; Seo, M. K. Full three-
dimensional power flow analysis of single-emitter–plasmonic-nano-
antenna system. *Opt. Express* 2015, 23, 11080–11091.
(5) Sawai, Y.; Takimoto, B.; Nabika, H.; Ajito, K.; Murakoshi, K.
Observation of a Small Number of Molecules at a Metal Nanogap
Arrayed on a Solid Surface Using Surface-Enhanced Raman Scattering.
*J. Am. Chem. Soc.* 2007, 129, 1658–1662.
(6) Huck, C.; Neubreich, F.; Vogt, J.; Toma, A.; Gerbert, D.;
Katzmann, J.; Hartling, T.; Pucci, A. Surface-Enhanced Infrared
Spectroscopy Using Nanometer-Sized Gaps. *ACS Nano* 2014, 8,
4908–4914.
(7) Ueno, K.; Juokazis, S.; Shibuya, T.; Yokota, Y.; Mizeliski, V.;
Sasaki, K.; Misawa, H. Nanoparticle Plasmon-Assisted Two-Photon
Polymerization Induced by Incoherent Excitation Source. J. Am. Chem. Soc. 2008, 130, 6928–6929.

(8) Kinkhabwala, A.; Yu, Z.; Fan, S.; Avlasevich, Y.; Millen, K.; Moerner, W. E. Large single-molecule fluorescence enhancements produced by a bowtie nanoantenna. Nat. Photonics 2009, 3, 654–657.

(9) Ming, T.; Chen, H.; Jiang, B.; Li, Q.; Wang, J. Plasmon-Controlled Fluorescence: Beyond the Intensity Enhancement. J. Phys. Chem. Lett. 2012, 3, 191–202.

(10) Tsuboi, Y.; Shoji, T.; Kitamura, N.; Takase, M.; Murakoshi, K.; Mizumoto, Y.; Ishihara, H. Optical Trapping of Quantum Dots Based on Gap-Mode-Excitation of Localized Surface Plasmon. J. Phys. Chem. Lett. 2010, 1, 2327–2333.

(11) Ureña, E. B.; Kreuzer, M. P.; Izhakhov, S.; Rigneault, H.; Quidant, R.; Oron, D.; Wenger, J. Excitation Enhancement of a Quantum Dot Coupled to a Plasmonic Antenna. Adv. Mater. 2012, 24, OP314–OP320.

(12) Jensen, R. A.; Huang, I. C.; Chen, O.; Choy, J. T.; Bischof, T. S.; Lonić, M.; Bawendi, M. G. Optical Trapping and Two-Photon Excitation of Colloidal Quantum Dots Using Bowtie Apertures. ACS Photonics 2016, 3, 423–427.

(13) Hentschel, M.; Metzger, B.; Knabe, B.; Buse, K.; Giessen, H. Linear and nonlinear optical properties of hybrid metallic–dielectric plasmonic nanoantennas. Beilstein J. Nanotechnol. 2016, 7, 111–120.

(14) Aouani, H.; Rahmani, M.; Navarro-Cía, M.; Maier, S. A. Third-harmonic-upconversion enhancement from a single semiconductor nanoparticle coupled to a plasmonic antenna. Nat. Nanotechnol. 2014, 9, 290–294.

(15) Schlather, A. E.; Large, N.; Urban, A. S.; Nordlander, P.; Halas, N. J. Near-Field-Mediated Plasmonic Coupling and Giant Rabi Splitting in Individual Metallic Dimers. Nano Lett. 2013, 13, 3281–3286.

(16) Santhosh, K.; Bitton, O.; Chantnoton, L.; Haran, G. Vacuum Rabi splitting in a plasmonic cavity at the single quantum emitter limit. Nat. Commun. 2016, 7, 11823.

(17) Chikkaraddy, R.; de Nijs, B.; Benz, F.; Barrow, S. J.; Scherman, O. A.; Rosta, E.; Demetriadou, A.; Fox, P.; Hess, O.; Baumberg, J. J. Single-molecule strong coupling at room temperature in plasmonic nanocavities. Nature 2016, 535, 127–130.

(18) Jeong, H.; Kim, D.; Xiang, D.; Lee, T. High-Yield Functional Molecular Electronic Devices. ACS Nano 2017, 11, 6511–6548.

(19) Wang, T.; Nijhuis, C. A. Molecular electronic plasmonics. Appl. Mater. Today 2016, 3, 73–86.

(20) Koenderink, A. F. Single-Photon Nanoantennas. ACS Photonics 2014, 7, 710–722.

(21) Galloway, C. M.; Kreuzer, M. P.; Acimović, S. S.; Volpe, G.; Correia, M.; Petersen, S. B.; Neves-Petersen, M. T.; Quidant, R. Plasmon-Assisted Delivery of Single Nano-Objects in an Optical Hot Spot. Nano Lett. 2013, 13, 4299–4304.

(22) Shoji, T.; Saitoh, J.; Kitamura, N.; Naganawa, F.; Murakoshi, K.; Yamauchi, H.; Ito, S.; Miyasaka, H.; Ishihara, H.; Tsuboi, Y. Permanent Fixing or Reversible Trapping and Release of DNA Micropatterns on a Gold Nanostructure Using Continuous-Wave or Femtosecond-Pulsed Near-Infrared Laser Light. J. Am. Chem. Soc. 2013, 135, 6643–6648.

(23) Geiselmann, M.; Marty, R.; Renger, J.; García de Abajo, F. J.; Quidant, R. Deterministic Optical-Near-Field-Assisted Positioning of Nitrogen-Vacancy Centers. Nano Lett. 2014, 14, 1520–1525.

(24) Jiang, M.; Kurvits, J. A.; Lu, Y.; Nurmikko, A. V.; Zia, R. Reusable Inorganic Templates for Electrostatic Self-Assembly of Individual Quantum Dots, Nanodiamonds, and Lanthanide-Doped Nanoparticles. Nano Lett. 2015, 15, 5010–5016.

(25) Arias-Zapata, J.; Cordeiro, J.; Böhme, S.; Girardot, C.; Garnier, J.; Bézard, P.; Nietuschkis, K.; Lontios, G.; Avgeropoulos, A.; Peyrade, D.; Zelmann, M. High throughput sub-10 nm metallic particles organization on templates made by block copolymer self-assembly and nanoimprint. Microelectron. Eng. 2015, 141, 155–159.

(26) Ndukaife, J. C.; Kildishev, A. V.; Nnanna, A. G. A.; Shalaev, V. M.; Wereley, S. T.; Boltasseva, A. Long-range and rapid transport of individual nano-objects by a hybrid electrothermoplasmonic nano-tweezer. Nat. Nanotechnol. 2016, 11, 53–59.