REVIEW

Light-induced manipulation with surface plasmons

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Abstract
We review recent advances achieved in the field of surface plasmon-based optical manipulations. We first discuss enhanced optical forces at the surface of a flat metal film and their use for self-organizing a large number of micro-objects. We then show how a suitable engineering of plasmon fields near micro-gold pads enables trapping at a specific location with much weaker laser intensity compared to conventional optical tweezers. This part is illustrated by a series of numerical simulations based on the theory of the Green dyadic. Finally, we show that, beyond their low power requirement, this new generation of integrated optical tweezers offers new perspectives in optical manipulation including parallel trapping with a single beam and controllable selectivity on the object polarizability.

Keywords: plasmon optics, near-field optics, optical manipulation

(Some figures in this article are in colour only in the electronic version)

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1. Introduction

The interaction of light with metal micro- and nano-structures supporting surface plasmon (SP) collective excitation has recently received considerable attention which has spread over different scientific communities, ranging from biology to integrated optics. In particular, enhanced and confined SP fields have been shown to be efficient for increasing light–matter interactions with small amounts of matter. They have successively been exploited for enhanced spectroscopy [1, 2], sensing of reduced numbers of molecules [3, 4] and to improve the efficiency of light sources and detectors [5, 6].

We show here how surface plasmon optics can successfully apply to optical manipulations in coplanar geometry. The latest advances of SP optics are exploited to engineer the optical near-field landscape at a patterned metal surface and achieve integrated 2D optical micro-traps. These novel SP traps have several advantages for integration over conventional 3D optical tweezers since they do not need any bulk optics and work with substantially reduced laser intensities. Furthermore, their coplanar geometry confers onto them unique properties such as their controllable selectivity to the specimen size.

After reviewing the current state of the art of optical manipulation based on evanescent fields in section 2, we discuss in section 3 enhanced optical forces and self-assembling of Mie colloids at a flat metal/dielectric interface supporting a surface plasmon. In section 4, we describe how a suitable plasmonic pattern can be designed to trap single colloids at determined positions of the surface. Finally, section 5 underlines the specificities of SP traps compared to conventional 3D tweezers and their implication on the trapping selectivity to different particle sizes.
2. State of the art in evanescent optical trapping

Conventional optical tweezers (OT), as first proposed by Ashkin in the 1970s [7] are implemented by tightly focusing, down to the diffraction limit, a laser beam with a high numerical aperture objective lens [8, 9]. An object located nearby the laser focus experiences two force contributions. The scattering force (or radiation pressure) is a repulsive force pointing along the incident \(k\)-vector while the gradient force, originating from the gradient of the electromagnetic field intensity, tends to pull it towards stronger intensity regions (figure 1(A)). The object gets stably trapped when the intensity profile at the laser focus enables the gradient force to dominate both scattering forces and Brownian motion. The total force magnitude depends on the trap profile and the electric polarizability of the object, determined by its dimensions and the dielectric function contrast with the surroundings. In practice, typical laser intensities used for trapping are of the order or greater than \(10^9\) W m\(^{-2}\). This way OT have been used for 3D manipulation of different kinds of systems immersed into a fluidic environment from micro-colloids and droplets [10, 11] to living cells [12–14].

Optical manipulations based on evanescent fields instead of propagating ones open new perspectives towards the integration of optical tweezers at the surface of a chip. Indeed, the intrinsic transverse confinement of surface fields enables maintaining the manipulated objects close to the surface.

In the simplest configuration, where a plane wave is totally reflected at an interface, the particle is pulled towards the surface by the gradient force and pushed by the scattering force (figure 1(B)) along the in-plane \(k\)-vector [15]. Using this concept, Kawata and co-workers first demonstrated optical guiding of micro- and nano-colloids at the surface of a prism illuminated under total internal reflection [16] and on top of a channel optical waveguide [17]. Recently, transport along optical waveguides has been implemented to achieve optical sorting of micro-colloids with different sizes [18]. Beyond in-plane guiding, trapping with evanescent fields requires additional in-plane field gradients necessary for maintaining the specimen at a fixed location. While an evanescent trap can be produced by using a TIR (total internal reflection) objective lens as reported in [19], several configurations overcoming the use of high NA objectives have been implemented. Large periodic arrays of evanescent traps formed by two interfering counter-propagating evanescent fields were reported by Cizmar et al [20, 21] (figure 1(C)). Similar results have been achieved by projecting a periodic diffractive element at the sample surface [22].

By exploiting recent concepts borrowed from nanooptics [23–29], one can use evanescent fields scattered by subwavelength objects (size \(\ll\) incident wavelength of light) to achieve non-diffraction-limited 3D optical field confinement potentially able to trap Rayleigh objects. In particular, light confinement at the extremity of a metallic tip [30, 31] (figure 1(F)) or at the output of a subwavelength hole in a metallic film [32, 33] (figure 1(E)) was predicted to enable optical trapping of very small objects down to a few tens of nanometers. The practical implementation of nano-trapping is associated with specific difficulties such as weak forces (due to small polarizability of particles with small dimensions) and strong Brownian motions which have until now prevented clear experimental demonstrations of these predictions.

Beyond conventional evanescent fields, surface plasmons (SPs) supported at metal/dielectric interfaces offer new opportunities in optical manipulation. SPs are resonant oscillations of the quasi-free electrons of a metal at its interface with a dielectric that are driven by a p-polarized electro-magnetic field. They lead to enhanced surface fields propagating along the interface and decaying exponentially away from it [34–36]. Because of their evanescent nature, SPs cannot be directly coupled to propagating light but require alternative configurations able to compensate the momentum mismatch with free space photons. In the most popular coupling scheme, known as the Kretchmann–Raether configuration, the metal/dielectric interface is illuminated under total internal reflection using a glass prism. In this way, efficient coupling to the SP mode, up to 90%, leads to a concentration of optical energy at the metal/dielectric interface responsible for a substantial enhancement of the incident field. This enhancement depends on the involved materials at the considered incident wavelength [34]. In the specific case of a gold/water interface, the theory predicts an intensity enhancement of several tens at 633 nm, expected to magnify both optical force components compared to conventional evanescent trapping [37].

In fact, the intrinsic properties of SPs could be of benefit to evanescent optical manipulation in two main manners.

- The enhancements of SP fields are expected to decrease the trapping intensity requirements.
- The ability of SP fields to be confined down to the subwavelength regime opens new perspectives to scale optical trapping down to the nanometer scale.

3. Enhanced optical forces and self-organization at a flat metal/dielectric interface

In a previous study [38], photonic force microscopy [39, 40] was used to measure quantitatively the enhanced optical forces experienced by a micro-object exposed to the SPs at a flat continuous metal/water interface. In good agreement with
theoretical predictions [37], experimental data revealed a strong enhancement factor (∼40) of the total force magnitude recorded near the plasmon resonance. In this section, we show how SP-induced optical forces enable self-organization under low laser intensity of a large number of free micro-beads at an extended flat metal surface [41].

The experimental set-up used for this experiment is sketched in figure 2. A thin gold layer (40 nm) is illuminated under total internal reflection by a slightly focused laser beam (∼50 μm diameter in the surface plane) under the plasmon resonant conditions (p-polarization, λ = 1064 nm, θ = 62°). The surface is exposed to a diluted solution of polystyrene (PS) micro-beads (5 μm) contained into a fluidic chamber of controllable thickness (10 μm). The dynamics of the colloids is monitored from the top using a long working distance objective connected to a camera.

Figure 3 shows the evolution with the incident laser power of the distribution of 5 μm PS beads at a flat gold/water interface when illuminated under the SP resonance conditions (p-polarization, λ = 1064 nm, θ = 62°). The ellipse show the approximative illumination area. From [41].

4.1. Numerical simulations

When patterning a metal surface to form finite metal pads, the boundaries confine the SP to the metal region and consequently reduce the spatial extension of enhanced SP fields. The resulting contrast between the enhanced field at the metal interface and the field at the bare substrate surface leads to an in-plane electromagnetic intensity gradient which can be engineered to form a stable potential well able to trap particles located in its vicinity.

To illustrate this concept, we start by computing the electric near-field intensity distribution nearby a gold pattern formed by several gold pads lying on a transparent surface and immersed in water. Let us consider the geometry shown in figure 4(A). In this configuration, a plane wave illuminates the system through the glass substrate under total internal reflection. We define E₀(r, ω₀) and S(r, r', ω₀) as the incident electric field and the field-susceptibility of the system in the absence of supported structure (this second-rank tensor is also called the Green dyadic function in the literature [27, 48]), respectively. One can show that the new electric field state (in the presence of the metal structures) is the solution of the Lippmann–Schwinger integral equation:

\[
E(r, \omega_0) = E_0(r, \omega_0) + \frac{\epsilon(\omega_0) - n_{\text{sur}}^2}{4\pi} \int S(r, r', \omega_0) \cdot E(r', \omega_0) \, dr',
\]

where \(\epsilon(\omega_0)\) represents the permittivity of the metal, \(n_{\text{sur}}\) the optical index of the surroundings, and the integral runs over the volume \(r\) occupied by the metallic structure.

In maps (B) and (C) of figure 4, two different incident polarizations are considered: p-polarization, where the incident electric field is contained in the incident plane, and s-polarization, where it is parallel to the glass/water interface. Each of the gold pads behaves as a dipolar polarizable object featuring different near-field optical responses depending on the polarization of the driving incident field. Under p-polarization, the dominant field component is perpendicular to the sample surface, leading to a field intensity maximum on top of each of the pads. Conversely, under s-polarization, the pads are polarized in the surface plane, leading to two maxima located just above their lateral edges. Finally, we remark that the disordered arrangement of the pads tends to downgrade the image–object relation even in the near-field zone.

A small object, located at position \(\mathbf{R}\), and exposed to a plasmonic near-field optical landscape, such as the one of figure 4, experiences an optical potential susceptible to
Figure 4. Example of simulations performed near disordered square-shaped gold pads embedded in water and supported by a plane transparent surface. The system is illuminated in total internal reflection: (A) top view of the model. The numerical method is described in [27]. (B)–(C) Maps of the normalized electric near-field intensity in p- and s-polarization, computed in a plane located at 35 nm from the top of gold pads (100 nm wide and 40 nm high). The surface wave is p-polarized in (B) where the scale varies from 0.65 to 5.2, and s-polarized in (C) where the scale increases from 0.1 to 5.3 (image size: 730 nm × 550 nm; incident wavelength 710 nm; and incident angle 70°).

Figure 5. Sketch of the force components experienced by an object near a finite plasmonic structure.

Figure 6. Optical binding energy maps computed by scanning a PS bead (80 nm in diameter) above a set of five gold disks (300 nm in diameter) embedded in water and supported by a plane glass surface. The sample is illuminated in total internal reflection under p-polarization. (A) Disordered arrangement (image size: 2250 nm × 1500 nm); the scale varies from −1.2 (dark) to −0.24 meV (bright). (B) Aligned arrangement (image size: 1800 nm × 1200 nm); the scale varies from −1.2 (dark) to −0.32 meV (bright). The incident wavelength and angle are 710 nm and 70°, respectively.

scattering force $F_{\text{scat}}$ pointing along the incident in-plane $k$-vector which will tend to guide the object along the surface. On the other hand, the field gradient near the gold structures creates attractive gradient forces $F_{\text{grad}}$, both in the plane and perpendicular to it, pulling the object towards the field intensity maxima [49]:

$$F_{\text{grad}} = -\nabla_R U_{\text{grad}}(R),$$

where the potential $U_{\text{grad}}(R)$ is a function of the optical indices $n_{\text{bead}}$ and $n_{\text{sur}}$ of the particle and surroundings, respectively, and the electric near-field intensity $|E(r, \omega_0)|^2$ is generated by the localized plasmons inside the bead according to

$$U_{\text{grad}}(R) = -\frac{n_{\text{bead}}^2 - n_{\text{sur}}^2}{16\pi} \int_V dv |E(r, \omega_0)|^2.$$
Figure 7. Vertical force maps (in fN) computed by scanning a polystyrene bead (40 nm in diameter) above single gold disks of different diameters $D$ embedded in water and supported by a plane transparent surface. The sample is illuminated in total internal reflection (image size: 1200 nm $\times$ 1200 nm). The incident wavelength and angle are 710 nm and 70°, respectively. The laser intensity is fixed at $10^9$ W m$^{-2}$.

(A) $s$-polarization and $D = 300$ nm; (B) $p$-polarization and $D = 300$ nm; (C) $s$-polarization and $D = 500$ nm; (D) $p$-polarization and $D = 500$ nm; (E) $s$-polarization and $D = 700$ nm; (F) $p$-polarization and $D = 700$ nm.

Gradient forces (associated to the depth of the potential well) on the one hand and the Brownian energy $k_B T$ (where $k_B$ and $T$ are the Boltzmann constant and the temperature, respectively) and the in-plane scattering force on the other hand. Our simulations for this geometry show that the restoring gradient forces dominate for a $p$-polarized incident surface wave and incident angles greater than 68°. In addition, as illustrated in figure 6, the well shape is highly sensitive to arrangement of the metal pads. For example, linear pad arrays produce thin energy channels that could be applied to the transport of a tiny amount of matter in coplanar geometry.

Figure 7 shows the distribution of the total normal force experienced by the bead as a function of the metal pad diameter and the incident polarization. This force component, $F_{\text{grad}Z}$, oriented along the axis perpendicular to the sample, characterizes the attraction of the bead by the plasmonic structure. For a given position $\mathbf{R} = (X, Y, Z = \text{cst})$ of the bead, it is computed by taking the first derivative of equation (3) and adding the normal component of the radiative force (see [50]). The regions where $F_{\text{grad}Z}$ takes significant values (in blue in figure 7) are rounded up by the circles that delimit the metallic disks. For all disk sizes, the trap
area appears to be significantly more confined in p-polarized than in s-polarized modes. Also, the force distribution gets more complex for increasing pad diameters due to retardation effects. Nevertheless, under p-polarization, all force maps are dominated by a sharp maximum at the forwards edge. We also observe, on the map (F) of figure 7, an enhancement by a factor 50 of the binding force $F_{\text{grad}}$ produced by the metal disk (with respect to its value away from the metal).

Making a simple analogy with 3D OT, similarly to the high NA objective lens in a conventional OT set-up, the metal disk somehow acts as a micro-lens that is able to provide from an extended illumination the in-plane field confinement necessary for trapping. This way, parallel trapping should also occur in an array of disks using the same homogeneous illumination as illustrated in figure 6.

4.2. Experimental results

Based on previous numerical predictions, experiments were carried out using the set-up of figure 2. Instead of a flat gold film, different patterns of micro-sized gold disks were fabricated using e-beam lithography combined with lift-off. In order to minimize the contribution of thermal-induced hydrodynamics, in addition to working with thin chamber thicknesses (<20 μm), we restricted ourselves to a low density of gold micro-disks.

Figure 8 summarizes, in a sequence of successive frames, a first experiment performed with an isolated gold disk (4.8 μm diameter and 40 nm height) located at the center of the screen and exposed to a solution of 4.88 μm PS beads. The sample surface is illuminated with a 710 nm laser beam near the plasmon resonance angle ($\theta = 66^\circ$). The illumination area and the incident power are 0.1 mm² and 250 mW, respectively, corresponding to an intensity $I \approx 2.5 \times 10^6$ W m⁻². Our measurements first show how the beads, away from the gold pad, are guided along a straight line from bottom to top. This is the scattering force (or radiation pressure) which pushes the beads at the glass surface along the incident in-plane $k$-vector. This movement is actually exploited to bring the beads towards the trapping area without using any fluidic flow. One of the guided beads passing upon the gold pad gets trapped and remains immobilized to a small portion of the disk as long as the illumination is maintained [47].

At this stage, one can get deeper insight into SP trap properties by analyzing the time-dependence position of the trapped bead and exploiting concepts of photonic force microscopy [38]. In order to maximize the accuracy of the measurement, we increase the total recording time by reducing as much as possible the scattering force magnitude. This leads us to work with an incident angle $\theta = 68^\circ$, where the contribution of the scattering force becomes weak while the restoring SP forces are maximum. Minimizing the scattering force also enables us to trap enough time under s-polarization and to assess the trap features when not coupling to the SP mode.

The tracking diagram of a 3.55 μm PS sphere trapped by a 4.8 μm gold disk under p-polarization (9 min acquisition time) is shown in figure 9(A). Already from these data it is interesting to see how the particle remains confined to a forward region of the pad as suggested by the numerical calculations of figure 6. This confinement is specific to p-polarization where the incident laser couples to the SP mode of the pad. In s-polarization the bead’s trajectory extends over nearly the whole pad area.
From the histogram of the bead positions, one can assess the shape of the potential function $U(l)$ ($l = X, Y$) it experiences. The Boltzmann distribution describes the probability density $r(l) = \exp(-U(l)/k_B T)/Z$, where $Z$ is the partition function normalizing the probability density function, $k_B$ the Boltzmann constant and $T$ the absolute temperature. This method permits us to check the assumption of a harmonic potential and when it is possible to proceed to the correlation [51] and the power spectral density analysis [52] of the position time-series for the evaluation of the trap stiffness as illustrated in figure 9. For the weak incident laser intensity used in our experiment, the potential depth in p-polarization is found to be greater than $4k_B T$. The resulting trap stiffness is about $30\text{ fN }\mu\text{m}^{-1}$ along $X$ and $17\text{ fN }\mu\text{m}^{-1}$ along $Y$. These values are one order of magnitude greater than under s-polarization [53].

To end this section, we now investigate experimentally the ability to achieve simultaneous parallel trapping in a pattern of several gold disks located within a homogeneous illumination area. Figure 10 shows parallel trapping of seven particles achieved in a hexagonal array of disks. Conceptually, the method can be extended to any type of pattern while the number of traps is limited by the size of the incident beam able to provide the minimum incident laser intensity necessary for trapping. However, it should be underlined that beyond a certain density of gold (i.e. for shorter distances between neighboring pads), agglomeration of beads occurs, disturbing their proper organization according to the gold pattern.

5. Tunability and selectivity of SP traps

After describing the concept of SP tweezers and their experimental implementation, we discuss in this last section their main physical specificities over conventional 3D tweezers and show how the latter can be exploited for engineering the trap and confer upon it a certain selectivity to the object polarizability.

Similarly to 3D tweezers, the ability to stably trap a nearby object is controlled by a right balance between (repulsive) scattering forces and (attractive) gradient forces. In conventional 3D tweezers, the scattering force points along the incident $k$-vector while gradient forces act both along the optical axis and perpendicular to it. Due to the 2D layout of SP tweezers, both contributions are in this case contained within the surface plane (see figure 5). The major difference though of SP tweezers over conventional 3D tweezers arises from the dependence of both scattering and gradient forces on the illumination parameters and in particular on the incident angle. On the one hand, the scattering force magnitude rises when decreasing the incident angle by improving the spatial overlap between the tail of the evanescent field and the object volume (see figure 11(A)). On the other hand, the strength of the gradient force follows the resonant behavior of the SP mode and reaches a maximum at the plasmon angle $\theta_{SP} = 68^\circ$ (see figure 11(B)). In fact the amount of energy coupled to the SP can also be accurately tuned by controlling the incident polarization, thus affecting the weight of the incident vertical field component. Consequently, the trapping properties of SP tweezers can be tuned by changing the balance between the two force components.

In order to evaluate the combined effect of the incident angle $\theta$ on the trapping properties of a single pad, we plot in figure 11(C) the trapping probability for a $4.88\mu\text{m}$ bead as a function of $\theta$. In the experiment, a probability of one has been attributed for trapping times longer than $120\text{ s}$. Applying this definition, three different regimes have been identified...
Figure 11. (A) Evolution with the incident angle of the guiding velocity of 3.55 and 4.88 μm PS beads at a glass/water interface. (B) Coupling efficiency to the SP at a flat 50 nm thick gold film. (C) Evolution with the incident angle of the trapping probability of a 4.88 μm PS bead near a 4.8 μm gold pad under both p- and s-polarization. (D) Illustration of the trapping mechanism for an incidence angle θ = 66°.

Figure 12. (A) Evolution with the incident angle of the trapping probability for both 3.55 and 4.88 μm PS beads under p-polarization. (B) Illustration of the selective mechanism (see video 2 at stacks.iop.org/JOptA/10/093001).

within the considered range of angles 64° < θ < 69° under p-polarization. While for θ < 65° the gradient restoring forces are weak since we stand out from the SP resonance and the scattering force is maximum, no trapping is achieved. Conversely, for θ > 67°, the local intensity at the pad surface is maximum while the scattering force is minimum, leading to an efficient trapping of the bead. The scattering force is actually so weak in this case that significant trapping is also observed under s-polarization without involving SPs. Beyond this similarity in the trapping probability, the effect on the trapping potential introduced by the polarization state has been discussed previously (figure 9). Between these two limit cases, there is a transient regime where the ratio between both force contributions varies sharply with θ. Within this range, we have identified incidence conditions for which the bead is systematically stably trapped under p-polarization while it quickly escapes under s-polarization. In this last case, the uncertainty in the time to release the bead arises from its Brownian dynamics.

Beyond the tunability of SP tweezers with the illumination parameters, one can additionally exploit the intrinsic dependence of both force components on the bead polarizability to control the trapping selectivity to the bead size observed in our prior works [47]. In order to illustrate this concept, we plot in figure 12 the trapping probability as a function of the incident angle (under p-polarization) for both 3.55 and 4.88 μm beads. Although for θ < 65° and θ > 68° both bead sizes behave similarly, between 66° and 67° the smaller particle is efficiently trapped while the bigger one is quickly released. We interpret this effect by remarking that the scattering contribution, two times stronger for the 4.88 μm bead (see figure 11), makes the balance between scattering and restoring forces opposite for the two bead sizes. This feature, specific to SP tweezers, offers in practice the possibility of controlling their selectivity to
determine objects out of a mix by simply adjusting the incident angle.

6. Conclusion

Optical manipulation based on surface plasmons is a novel and promising application of plasmonics which opens new opportunities for the elaboration of future integrated optofluidic devices. By exploiting the enhanced optical forces at homogeneous and patterned metal surfaces, self-organization and parallel trapping is achieved under weak laser intensity compared to conventional optical tweezers. Interestingly, the different force contributions involved in SP tweezers can be tuned by adjusting the illumination parameters, permitting dynamic control of their trapping ability and selectivity on the object polarizability.

While the method is well controlled for the manipulation of micrometer-sized objects, the future challenge is in extending SP-based manipulation to objects much smaller than the incident wavelength. Among the possible strategies, it was recently suggested to exploit the electromagnetic coupling between metal nanoparticles [46, 54].

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References


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