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Optical Trapping and Manipulation of Nano-objects with an Apertureless Probe

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We propose a novel way to trap and manipulate nano-objects above a dielectric substrate using an apertureless near-field probe. A combination of evanescent illumination and light scattering at the probe apex is used to shape the optical field into a localized, three dimensional optical trap. We use the coupled-dipole method and the Maxwell stress tensor to provide a self-consistent description of the optical force, including retardation and the influence of the substrate. We show that small objects can be selectively captured and manipulated under realistic conditions.

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Since the seminal work of Ashkin on radiation pressure, the possibility to exploit the mechanical action of optical fields to trap and manipulate neutral particles has spawned a wide range of applications. From atomic and nonlinear physics to biology, optical forces have provided a convenient way to control the dynamics of small particles (see Ref.[1] for a review). Optical tweezers, for example, have proved useful not only for trapping particles, but also for assembling objects ranging from microspheres to biological cells [2]. However, most of these manipulations involve particles whose size is between one and several micrometers. While for much smaller particles, such as atoms or molecules, the scanning tunneling microscope provides a powerful tool for manipulation and engineering [3], dealing with neutral particles of a few nanometers requires new experimental approaches.

The idea of using a metallic probe to trap small particles was reported by Novotny et al [4]. Their calculations showed that strong field enhancement from light scattering at a gold tip could generate a trapping potential deep enough to overcome Brownian motion and capture a nanometric particle in water (a related work by Okamoto and Kawata demonstrates theoretically the trapping of a nanometric sphere in water near an aperture probe [5]). This technique should be delicate to implement in practice for three reasons. First, before the particle can be captured, Brownian fluctuations will have a disruptive effect. Second, radiation pressure from the illuminating laser will impart momentum to the particle. Therefore, one would have to capture a moving object. Finally, it will be rather difficult to use a near-field probe to find in water a particle a few nanometers in size. One might wait for a particle to wander in the trapping region, but such an operating mode does not allow for selective capture.

In this letter, we propose an experimental scheme to selectively capture and manipulate nanoparticles in vacuum or air above a substrate, using the tungsten probe of an apertureless near-field microscope. The particles are not in a liquid environment, hence there is no Brownian

motion and the apertureless probe can be used as a near-field optical probe to localize and select the particles [6,7]. This is an important asset when different particles have to be placed according to a specific pattern or when interactions between particles are investigated. For example, with our configuration quantum-dot nanocrystals (CdSe/ZnS nanospheres a few nanometers in diameter) could be placed in a specific geometry to study dot-dot interactions, or one nanocrystal could be isolated to study single-dot properties [8].

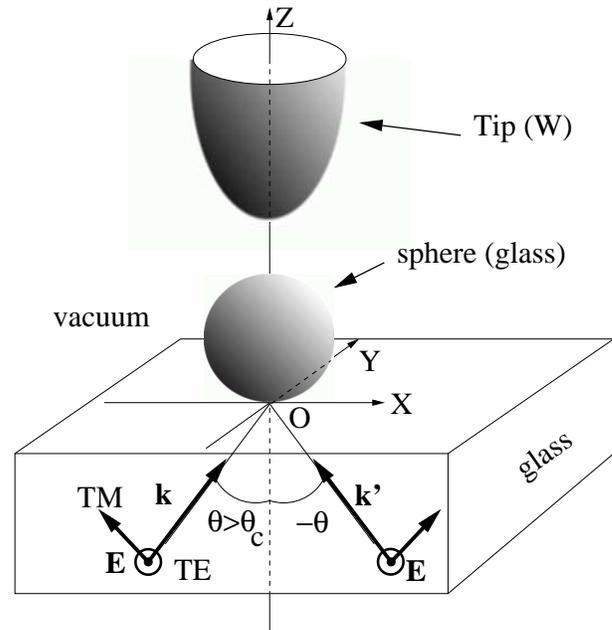


FIG. 1. Scheme of the configuration. A dielectric sphere (radius 10 nm) on a flat dielectric surface is illuminated under total internal reflection. A tungsten probe is used to create an optical trap.

We first consider a spherical particle with radius 10 nm placed in air on a substrate (Fig. 1). Unless it is stated otherwise the permittivity of both the particle and the

substrate is $\epsilon = 2.25$. The particle is illuminated (wavelength 500 nm) by two counter-propagating evanescent waves created by the total internal reflection of plane waves at the substrate/air interface (angle $\theta > \theta_c = 41.8^\circ$ with $\sqrt{\epsilon} \sin \theta_c = 1$). These two waves have the same polarization and a random phase relation. [9] This symmetric illumination ensures the lateral force vanishes when the sphere is just below the tip, thus avoiding that the sphere be pushed away from the tip.

We study the interactions between the particle and a tungsten probe (commonly used in apertureless microscopy) with a radius at the apex of 10 nm. The theory used to compute the optical forces has been presented in detail elsewhere [10]. Here we give a succinct summary. We use the coupled-dipole method [11,12] to model the light scattering and find the electromagnetic field inside the tip and the particle. Note that this procedure takes into account the multiple scattering between the sphere, the tungsten tip, and the substrate. We then use the Maxwell stress tensor technique to derive the optical forces [13]. We emphasize that the stress tensor approach is exact and does not rely on any assumption regarding the nature of the field (whether evanescent or propagating) or of the objects.

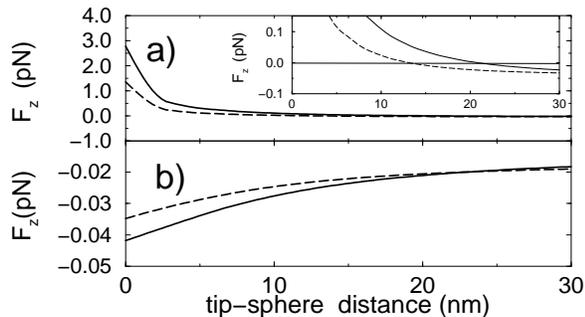


FIG. 2. z component of the force experienced by the sphere versus the distance between the tip and the sphere. Solid line : $\theta = 43^\circ$. Dashed line : $\theta = 50^\circ$. a) TM polarization. b) TE polarization.

All forces are computed for an irradiance of $0.05 \text{ W}/\mu\text{m}^2$ (this corresponds, for example, to a 5W laser beam focused over an area of $100 \mu\text{m}^2$). Figure 2 shows the z component of the force experienced by the sphere versus the vertical position of the tip above the sphere, for both TE and TM polarizations and for two angles of illumination ($\theta = 43^\circ$ and $\theta = 50^\circ$). As the tip moves closer to the sphere, the evolution of the force for the two polarizations is radically different. For TM illumination there is a large enhancement of the field near the apex of the probe because of the discontinuity of the z component of the electric field across the air/tungsten boundary [4]. This enhancement is responsible for the force being positive at short distances. Note that for the sphere, the

force of gravity is on the order of 10^{-7} pN . The z component of the force experienced by the sphere (when the tip is in contact with the sphere) is about 3pN which is 10^8 times its weight. Hence, gravity can be neglected. Fig. 2a shows that for TM polarization the force is larger for $\theta = 43^\circ$ than for $\theta = 50^\circ$. This is related to the slower decay of the evanescent wave for the smaller angle, which results in a weaker coupling between the sphere and the substrate. Moreover, the slower the decay of the evanescent field, the larger the field that reaches the tip, and the larger the (positive) gradient force caused by the field enhancement at the apex of the probe. As a consequence, when the tip approaches the sphere, the sign reversal (negative to positive) of the z component of the force occurs at a larger distance for $\theta = 43^\circ$ ($z=21 \text{ nm}$) than for $\theta = 50^\circ$ ($z=13 \text{ nm}$). This is shown in the inset of Fig. 2a. On the other hand, for TE polarization (Fig. 2b), the magnitude of the z component of the force increases while the force remains negative (directed toward the substrate and away from the tip) as the tip gets closer to the particle. This prevents any trapping. Our calculations show that this behavior is caused by the decrease of the field inside the upper part of the sphere which, in turn, causes a decrease of the gradient force. Another way of explaining this is to note that because the apex of the tip and the sphere are small compared to the wavelength, they can be approximated by two dipoles. For TE polarization these dipoles are essentially parallel to the substrate and as shown in Ref.[14], two parallel dipoles tend to repel each other. Since the magnitude of the dipole is stronger for $\theta = 43^\circ$ (due to the larger intensity of the field), the repulsive force is also stronger. Note that for a silver tip at the plasmon resonance frequency, the force acting on the sphere is positive for both polarizations. At the frequency considered here, tungsten behaves like an absorbing dielectric and the force is positive only for TM polarization.

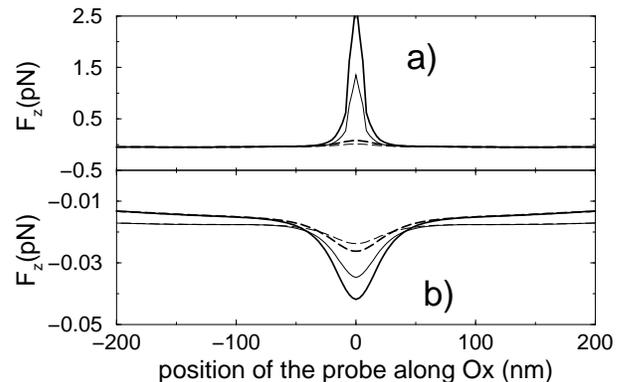


FIG. 3. Force along z experienced by the sphere as a function of the lateral position of the probe. The sphere is at the origin. a) TM polarization, b) TE polarization. Thick lines : $\theta = 43^\circ$. Thin lines : $\theta = 50^\circ$. The tip is either 20 nm (solid lines) or 31 nm (dashed lines) above the substrate.

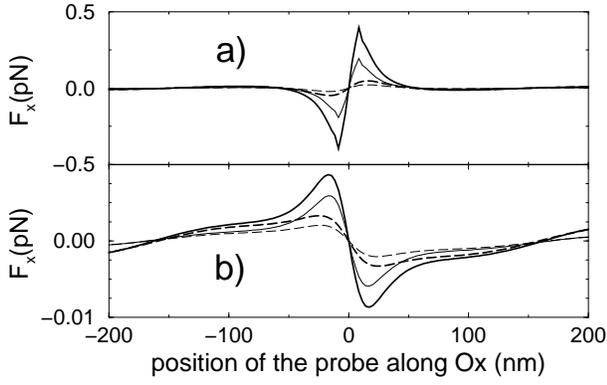


FIG. 4. Same as Fig. 3 for the force along x .

To fully assess the probe-particle coupling we study the evolution of the force experienced by the particle as the probe is moved laterally, the sphere remaining fixed. The components of the force acting on the sphere are plotted in Figs. 3 and 4 for two distances between the tip and the substrate (20 nm and 31 nm), and for two angles of illumination (43° and 50°). For TM polarization (Fig. 3a), the z component of the force is negative when the tip is far from the particle. When the tip gets closer, the particle starts experiencing a positive force along z . The change of sign occurs between $|x| = 30$ nm for $\theta = 43^\circ$ and $z = 20$ nm, and $|x| = 7$ nm for $\theta = 50^\circ$ and $z = 31$ nm. For TE polarization the force is negative (Fig. 3b). Similarly, we plot in Fig. 4 the lateral force. The symmetry of the force plot is a consequence of the symmetric illumination. For TM polarization (Fig. 4a) this force tends to push the particle toward the tip. For example, if the tip is located at $x = 10$ nm, we can see in Fig. 4a that the x component of the force experienced by the sphere is positive, hence the lateral force pushes the sphere toward the tip. If the tip is located at $x = -10$ nm, the x component of the force is negative and, again, it pushes the sphere toward the tip. Therefore, when the tip and the particle are close enough for the z component of the force to be positive and to lift the particle off the surface, the lateral force actually helps bringing the particle in the trap. Again TE polarization gives a different result. The lateral force always pushes the particle away from the tip. However, since the magnitude of the (downward) z component of the force is larger than the x component by a factor of 5, we expect that the sphere is not displaced when the tip is scanned over it under TE illumination. Note that apertureless probes are often used in tapping mode when imaging a surface. This mode minimizes the lateral motion imparted to the object by the optical force.

We have shown that a tungsten probe can be used to trap efficiently a nanometric object above a surface using TM illumination. For nanomanipulation it is important to assess the stability of the trap as the probe lifts the particle off the substrate. Figure 5 shows the z compo-

nent of the force when the sphere is fixed at the apex of the tip and the tip is moved vertically (solid and dashed curves). The optical force remains larger than the weight of the particle over a range of several tens of nanometers. The particle can therefore be manipulated vertically as well as horizontally. Note that the evolution of the force with the distance to the substrate is linear rather than exponential. The particle experience a negative gradient force due the exponential decay of the intensity of the illumination. At the same time, the particle suffers a positive gradient force due to the field enhancement at the tip apex, which also decreases exponentially with z because this enhancement depends on the intensity of the evanescent illuminating light. The competition between these two contributions results in an weak decrease of the trapping force as the particle is moved away from the substrate. If we change the nature, size or shape of the object the magnitude of the force changes but the conclusions are qualitatively the same (Fig. 5, curves with symbols).

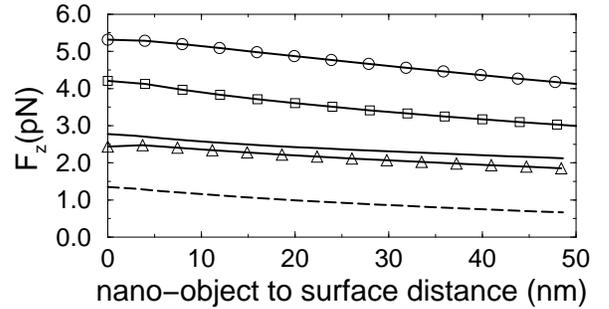


FIG. 5. z component of the force experienced by a particle as a function of the distance between the particle and the substrate under TM illumination. The particle is placed at the apex of the probe. Solid line : sphere ($\epsilon = 2.25$, radius: 10 nm, $\theta = 43^\circ$). Dashed line: sphere ($\epsilon = 2.25$, radius: 10 nm, $\theta = 50^\circ$). Circles : sphere ($\epsilon = 2.25$, radius: 30 nm, $\theta = 50^\circ$). Triangles (force $\times 0.1$): sphere of gold ($\epsilon = -2.81 + 3.18i$, radius: 10 nm, $\theta = 50^\circ$). Squares : cube ($\epsilon = 2.25$, size: 20 nm, $\theta = 50^\circ$).

It is fundamental to know whether our procedure also works if several particles are clustered together. We consider a set of three spheres (radius 10 nm, permittivity 2.25) aligned along x . The probe is placed above the middle sphere. We account for the multiple scattering between the three spheres, the substrate and the tip, therefore the optical binding experienced by the spheres [14] is included in our description. For this configuration, TE illumination again does not lead to trapping. For TM illumination, we plot in Fig. 6 the z component of the force experienced by the middle sphere and by those on the sides as a function of the vertical distance between the probe and the middle sphere. For an angle of incidence $\theta = 43^\circ$, the z component of the force, although largest

for the middle sphere, is positive for all the spheres. This could be a problem if one wanted to manipulate only one particle. The central particle can be selectively trapped by increasing the angle of incidence of the illuminating beams to tighten the trap in the x direction (Fig. 4). In Fig. 6 we see that for $\theta = 50^\circ$ the optical force induced by the probe is positive only for the middle sphere. This remains true for three spheres aligned along y . Figure 7 shows the extraction of the middle sphere by the tip. Our calculation shows that the vertical force experienced by the two side spheres remains negative when the probe moves away from the substrate. Therefore, the spheres on the sides do not hinder the capture of the middle sphere.

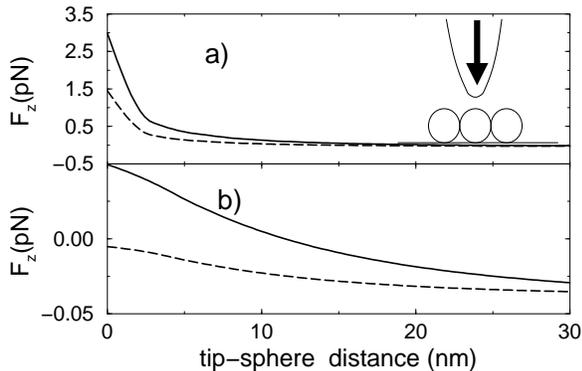


FIG. 6. Force along z versus vertical position of the probe for three spheres aligned along x . The probe is centered over the middle sphere. Solid line $\theta = 43^\circ$. Dashed line $\theta = 50^\circ$. a) Force on the middle sphere. b) Force experienced by the spheres on the sides.

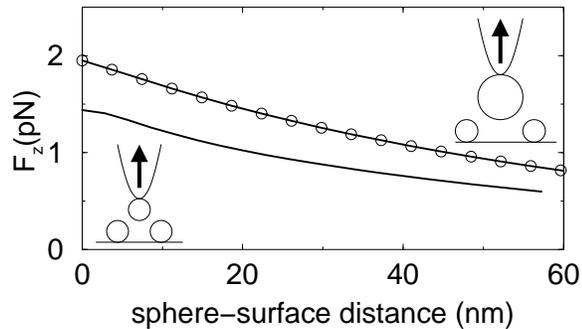


FIG. 7. Force along z as experienced by the middle sphere (solid line) versus vertical position of the middle sphere trapped at the apex probe ($\theta = 50^\circ$). Solid line : sphere with a 10 nm radius. Circles : sphere with a 20 nm radius.

In Fig. 7, the curve with circles shows that the selective trapping works as well when the middle sphere is twice as big as the others. Once the chosen particle has

been trapped, it can be moved above the substrate. Our calculations show that the presence of other particles on the substrate does not destroy the trap, provided that during the manipulation the trapped sphere is kept at least 5 nm above the spheres that are on the substrate. We have checked that if the optical binding causes the side spheres to move laterally toward the middle sphere, increasing the angle of illumination still creates a negative (downward) force on the two side spheres, and the selective capture of the middle sphere is not hampered.

The procedure to trap a small object with a tungsten tip is therefore the following: first TE illumination is used while the tip scans the surface in tapping mode or in constant-height mode if the area under investigation is small enough. Such modes avoid the displacement of the particle by the tip. Once an object has been selected, the probe is placed above the object and the polarization of the illumination is rotated to TM. The probe is then brought down over the particle and captures it. The probe can then move the particle above the substrate, both horizontally and vertically, to a new position where it can be released by switching back to TE polarization (note that if, for some reason, one wishes to move the particle over distances larger than the size of the illumination spot, one could move *the sample* with piezoelectric device once the sphere is trapped at the apex of the tip). The lack of trapping under TE illumination is actually an important advantage during the imaging (selection) and release phases of the manipulation. Indeed, under TE illumination, when the tip is above a particle it actually increases the downward optical force, which contributes to prevent the tip from sweeping the particle away.

In conclusion, we propose a new method to trap and manipulate nanometric particles in air above a substrate, using an apertureless tungsten probe. The probe is used to scatter two counter-propagating evanescent waves, generating an optical trap at the apex. We showed that an object of a few nanometers, can be selectively trapped and manipulated. An interesting extension of this work will be a study of the influence of different illuminations (e.g. focused beam) and a systematic study of the influence of the nature of the particle and the tip on the trapping mechanism. For example, the strong spectral dependence of the electromagnetic response of metal particles (or resonance excitation of both dielectric and metallic particles) could lead to a material selective trapping.

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