Photo-induced force mapping of plasmonic nanostructures

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Abstract:

The ability to image the optical near-fields of nanoscale structures, map their morphology and concurrently obtain spectroscopic information, all with high spatiotemporal resolution, is a highly sought-after technique in nanophotonics. As a step towards this goal, we demonstrate the mapping of electromagnetic forces between a nanoscale tip and an optically excited sample consisting of plasmonic nanostructures, with an imaging platform based on atomic force microscopy. We present the first detailed joint experimental-theoretical study of this type of photo-induced force microscopy. We show that the enhancement of near-field optical forces in gold disk dimers and nanorods follows the expected plasmonic field enhancements, with strong polarization sensitivity. We then introduce a new way to evaluate optically-induced tip-sample forces by simulating realistic geometries of the tip and sample. We decompose the calculated forces into in-plane and out-of-plane components and compare the calculated and measured force enhancements in the fabricated plasmonic structures. Finally, we show the usefulness of photo-induced force mapping for characterizing the heterogeneity of near-field enhancements in precisely e-beam fabricated nominally alike nanostructures - a capability of widespread interest for precise nanomanufacturing, SERS and photocatalysis applications.

KEYWORDS: Plasmonics, near-field scanning optical microscopy, gradient force, photocatalysis, nanocharacterization

Nanoscale optical materials such as photonic crystals¹, plasmonic materials^{2,3}, metamaterials^{4,5} and two-dimensional materials^{6,7,17}, have revolutionized the field of nanophotonics serving as ideal platforms to study and control light-matter interactions. Consequently, such materials have opened up an array of possibilities and applications in biomedicine^{8,9}, photonic circuitry^{10–12}, catalysis^{13–19}, sensing^{20,21}, and photovoltaics²². The optical properties of such designer materials are strongly dependent on the arrangement and morphology of metallic and dielectric elements that make up the bulk structures^{23,24}. Thus, it becomes important to visualize and image not only the topography and structural make-up of nanoscale optical materials, but also their electromagnetic interactions. Furthermore, certain chemical and biological applications call for the spectroscopic understanding of photo-induced mechanisms^{25,26}. *The correlation between morphology and near-field optical properties is especially strong for plasmonic particles, since small changes in morphology can significantly modify local and nonlocal near-field optical interactions*^{27,28}. Thus, it becomes vital to simultaneously image the morphology and near-field environment of such nanoscale structures, along with the ability to perform physicochemical spectroscopy with a high spatiotemporal resolution and selectivity.

As a first step towards this goal, we present experimental measurements where the photo-induced forces and the topography of plasmonic model systems are recorded simultaneously. Equally important, this is the first joint experimental-theoretical study of this type of photo-induced force microscopy. While past modeling work often simplified photo-induced forces between the tip and the sample by a dipole-dipole model or a two-sphere model we have extended these works by modeling realistic geometries of both, the AFM tip and the sample similar to the ones used in the experiment. Plasmonic nanostructures with their strong field

enhancements serve as ideal model system for detailed comparison between experimental and theoretical investigations.

Techniques such as atomic force microscopy (AFM), scanning electron microscopy (SEM) and transmission electron microscopy (TEM) for example, enable the imaging of physical properties of nanoscale materials with high spatial resolutions. Other methods such as near-field scanning optical microscopy (NSOM)^{26,29,30}, scanning photoionization microscopy (SPIM)^{31–33}, electron energy loss spectroscopy (EELS)^{26,30} and second-harmonic generation microscopy³⁰, have been used as tools to image the optical near-field beyond the diffraction limit. Nanoscale optical properties can be mapped with a spatial resolution of ~ 10 nm with a NSOM, but the signals collected through this technique typically also possess a strong background noise from parasitic scattering, requiring image deconvolution techniques to circumvent this problem³⁰. While electron microscopy-based techniques such as EELS fare better in terms of spatial resolution, they come with other drawbacks such as lack of control over polarization for probing samples and limitations in the range of samples that can be analyzed³⁰.

Several strategies have been explored to allow for the simultaneous imaging of topography and the electromagnetic near-field environment of nanoscale materials, with high resolution and spectral selectivity. Tip-based microscopies allow for *indirect* imaging of the near-field of nanoscale materials by probing photo-induced changes in chemical, physical or spectroscopic signatures through a nanoscale tip. One example of such an indirect imaging technique, is infrared absorption AFM, where the sample is excited with infrared light and the resulting thermal expansion of the sample is probed with an AFM tip^{34,35}. Alternatively, optical near-fields of nanostructures have also been imaged by monitoring the changes in the topography of a spin-coated polymer due to photoinduced chemical effects³⁶, and by functionalizing the AFM tip with

quantum dots, in order to provide a contrast mechanism for the imaging of electric field enhancements³⁷, the study of charge transfer dynamics³⁸ and resonant energy transfer microscopy³⁹. Another *indirect* approach to imaging the optical near-field environment is to track light-induced nanoscale forces using tip-based detection schemes. Although the nature of optical forces depends on several factors, such as tip-sample distance and the dielectric response of the sample, the idea of mapping such forces has been conceptually demonstrated in several systems. These include the imaging of the optical near-field of metallic and dielectric nanoantennas and nanoparticles^{40–43}, detection of molecular resonances⁴⁴, measurement of Casimir forces⁴⁵, the observation of extraordinary optical momentum and force of light⁴⁶, measurement of the optical force induced by a plasmonic cavity⁴⁷, and the identification of chemical components in block copolymers⁴⁸.

In this work, we utilize a technique, termed as photo-induced force microscopy (PiFM)^{42–44,48}, where a nanoscale tip is brought into the vicinity of an optically excited sample. As a result, the photo-induced forces between the tip and the sample can be detected (at the second order resonance frequency of the tip) with nanometer-scale spatial resolution, along with topographical information (at the first order resonance) that is simultaneously acquired with the atomic force microscopy channel.

We apply this technique to map the photo-induced force intensities in gold nanorods and disk dimer structures at their longitudinal plasmon resonance wavelengths where field enhancements at the tips and junctions will be large. We show that the force detection technique presented here qualitatively follows the expected polarization-dependent plasmonic field enhancements. We introduce a new way to evaluate the optically induced tip-sample forces theoretically by considering realistic geometries of the tip and sample, rather than simplifying the geometry of

the tip, as is done conventionally. We also decompose the calculated force into its in-plane and out-of-plane components and compare them with the experimental signals. We show that the enhancements in the measured photo-induced forces corresponding to plasmonic enhancements compare relatively well with the calculated photo-induced force enhancements. Finally, we explore the effect of an offset in alignment between the tip and the center of the focused beam spot on the symmetry of the force enhancement map.

In our experiments, we utilized a recently introduced commercial AFM platform (Vistascope from Molecular Vista, inc.). The experimental setup consists of a sample stage scanner, and a scan head to hold the cantilever (Fig. 1). Unlike traditional AFM schemes, the sample is positioned on top of an oil-immersion objective with a NA = 1.4 (PlanApo, Olympus). The excitation source is a continuous wave laser (OBIS 785 LX, Coherent, Inc.), from which, light with a wavelength $\lambda = 785$ nm is guided onto the back aperture of the oil-immersion objective. The laser beam overfills the back aperture and generates a tight focal spot on the sample plane. The average intensity at the focal plane was ~ 0.34 mW/ μm^2 . The diameter of the focused spot on glass, determined by raster scanning the objective, was ~ 0.86 μm (supplementary section). A linear polarizer placed in the optical path, was used to control the polarization of the incident light. The topographic information was collected at the first order resonance frequency of the cantilever, f_0 , and the force signal was simultaneously collected at the second order resonance frequency, f_1 . The excitation laser was digitally modulated at the sideband frequency $f_m = f_1 - f_0$ (Fig. 1). Tapping mode AFM is known to be more sensitive to the out-of-plane component of the optical force, thereby reducing noise from the longer range, in-plane component^{43,49}. We used a gold-coated silicon tip (TAP300GB-G, Budget Sensors) with tip radius ~ 25 nm, force constant =

40 N/m and the first and second order resonance frequencies, $f_0 \sim 270$ kHz and $f_1 \sim 1678$ Hz, respectively.

It is well known that metallic nanostructures can support plasmon resonances that can induce strong localized electric fields^{22,50}. Hence, we chose to work with gold nanorods, which exhibit symmetric field enhancements at the tips of the rods^{51,52}, and gold disk dimers, which can support strong field localization in the dimer gap^{53–56}, when excited with longitudinally polarized light. Using FDTD simulations, we optimized the gold nanorods to be 110 nm in length and 25 nm wide. The gold dimers were designed to be 150 nm in diameter each, with a 30 nm gap. Both structures were ~ 35 nm thick and fabricated by electron beam lithography on glass cover slips (Figure 1).

At an incident wavelength of $\lambda = 785$ nm, FDTD simulations reveal a strong enhancement in the electric field ($|E|$) at the gap of the disk dimers when light is polarized along the dimer axis (Figure 2a). For perpendicular polarization, the longitudinal gap mode is not excited, and the maximal field enhancement is ~ 2.5 times weaker and located on the side of the disks rather than in the junction. Figure 2b shows the experimentally measured photo-induced force intensity maps for both polarizations. For the case of longitudinal polarization, the strong plasmonic enhancement at the dimer gap is readily apparent in the force images. Similarly, for transverse polarization, the force enhancement follows the expected electric field enhancement predicted by FDTD simulations, where the gap mode vanishes. The maximal photo-induced force enhancement in the case of longitudinal polarization is stronger by ~ 2.1 times in comparison with the transverse case, in good agreement with the calculated near-field electric field enhancements in our plasmonic structures (Figure 2a). Figure 2c shows a representative topography map of the fabricated gold disk dimer structure, corresponding to the same structure

from the force intensity image shown in the bottom panel of Figure 2b. From the topography image, the thickness of the structure is ~ 38 nm, the full-width at half maximum of an individual disk is ~ 147 nm and the dimer gap is ~ 32 nm.

Although we observe relatively good agreement between the calculated total electric field (2.5x) and measured optical force (2.1x) enhancements when comparing illumination with the two orthogonal polarizations, one generally has to be careful in making quantitative comparisons between the electric field and optical forces. In typical tip-based measurements of the near-field environment of plasmonic structures, the measured electric fields would be convolved with the tip, with a strong dependence on its geometry and material, leading to discrepancies with rudimentary predictions^{57,58}. Moreover, the correlation between electric fields and photo-induced tip-sample forces is a non-trivial one, as we discuss below. Thus, a better way to make quantitative comparisons between experiments and calculations is to evaluate the photo-induced forces by considering the real geometry and materials of the AFM tip and sample and evaluating both the in- and out-of-plane components of the calculated forces and comparing them to the measured signals.

To calculate the tip-sample optical forces, most studies so far have treated the tip and sample as point dipoles^{42,43}, a reasonable approximation when their dimensions are much smaller than the wavelength of the incident light. However, such an approximation may fail when we investigate plasmonic structures excited by light of a wavelength comparable to their size. One approach to circumvent this issue is to numerically calculate the field induced by the samples and still treat the tip as a nonperturbative point dipole, which will not change the field distribution. We also assume that the tip has no static dipole moment. In this case, the induced dipole moment is

proportional to the electric field and the cycle-averaged optical force experienced by the tip may be approximated as⁵⁹

$$\langle \mathbf{F} \rangle = \frac{\alpha_t'}{2} \nabla \langle |\mathbf{E}_p|^2 \rangle + \omega \alpha_t'' \langle \mathbf{E}_p \times \mathbf{B}_p \rangle, \quad (1)$$

where $\mathbf{E}_p$ and $\mathbf{B}_p$ are the electric and magnetic field components induced by the nanoparticle, respectively, ω is the angular frequency of light. In this expression, α_t denotes the complex polarizability of the tip with real and imaginary components $\alpha_t = \alpha_t' + i\alpha_t''$. Note that Eq. 1 is derived⁵⁹ in the dipolar limit, the linear regime and the non-relativistic limit, assuming the velocity of the center of mass is small compared to the speed of light c , for an arbitrary monochromatic wave with harmonic time-dependence, but with the restriction that the phase needs to vary spatially much stronger than the amplitude. Furthermore, the inhomogeneity of magnetic field is neglected and it is assumed that the polarizability α_t is spatially invariant. The polarizability α_t can be estimated using the Clausius-Mossotti equation

$$\alpha = 4\pi\epsilon_0 \frac{\epsilon-1}{\epsilon+1} a^3, \quad (2)$$

where ϵ is the dielectric permittivity of the sphere and a is the radius. Because we use a tip that has a radius of curvature ~ 25 nm and height ~ 17 μ m, it is not appropriate to treat it as a point dipole, since the field inside the tip is nonuniform and will change the field distribution around the plasmonic nanostructure. To get a more accurate estimate of the optical force experienced by the tip, we use a more general method by calculating the surface integral of the Maxwell stress tensor^{43,59,60}, from which, the force experienced by the tip is given by

$$\langle \mathbf{F} \rangle = \int_S \langle \vec{\mathbf{T}}(\mathbf{r}, t) \rangle \cdot \mathbf{n}(\mathbf{r}) ds, \quad (3)$$

where $\mathcal{S}$ is an arbitrary surface enclosing the tip, $\mathbf{n}$ is the unit vector perpendicular to the surface and $\vec{T}(\mathbf{r}, t)$ is the Maxwell stress tensor, expressed in the fields as

$$\vec{T} = \epsilon_0 \epsilon \mathbf{E} \mathbf{E} - \mu_0 \mu \mathbf{H} \mathbf{H} - \frac{1}{2} (\epsilon_0 \epsilon E^2 + \mu_0 \mu H^2) \vec{I}. \quad (4)$$

At this time, it is computationally too expensive in terms of processor memory to include the geometry of a realistically dimensioned tip that is 17 μm long in the simulation box. Therefore, most studies treat the tip as a small spherical particle and calculate the optical force experienced by this particle using Eq. (3)^{42,61}. Although such a simplification enables one to approximate the spatial variation of optical forces relatively well, it underestimates the amplitude of the z-component of the optical force in plasmonic environments with strong field gradients. It is therefore important to model the tip with realistic geometry and dimensions. To achieve this, we propose a new quantity, the *induced optical force*, defined as $F_{\text{induced}} = F_{\text{with sample}} - F_{\text{without sample}}$, where $F_{\text{with sample}}$ is the optical force around the sample and $F_{\text{without sample}}$ is the optical force at the same position but in the absence of the sample. This quantity is more reasonable than the absolute value of $F_{\text{with sample}}$ itself since the PiFM images are using the signal detected from the place without samples as the reference. Considering that the field distribution around the upper part of the tip does not change significantly in absence of the sample, we can calculate F_{induced} using Eq. 3, by integrating over an unclosed integration box where the upper surface is left “open” (Figure 3a). We should thus be able to obtain an accurate estimate of F_{induced} using this method, as long as the box height is sufficiently large.

To determine the minimum size of the integration box that can be used to evaluate the tip-sample forces with sufficient accuracy, we start by examining the optical force around a single Au rod with length = 110 nm and width = 30 nm, illuminated by a cw light source at a wavelength $\lambda =$

785 nm (Figure 3a). The finite difference time domain (FDTD) method (Lumerical Solutions) is used to calculate the electric and magnetic field components in vicinity of the tip. We consider a Gaussian beam with a $(1/e^2)$ diameter ~ 800 nm, determined by the diameter of the right lobe of the E_z^2 field distribution (Supporting Figure S1). This beam is focused at the center of the tip, with amplitude $= 2.6 \times 10^5$ V/m, incident through an oil-immersion objective with NA = 1.4, which corresponds to an average laser intensity of $I_0 = 0.34$ mW/ μm^2 , similar to our experimental parameters. We model the tip as a silicon cone covered by a 70 nm thick gold film, with a radius of curvature = 20 nm and a cone angle = 20° , similar to the geometry of the tip used in experiments. The tip is positioned at a distance of 5 nm above the gold nanorod and an unclosed integration box with height h (Figure 3a). As can be seen in Figure 3a, F_{induced} is well converged around $h \sim 500$ nm.

Next, we compared the efficacy of our force evaluation method (F_{induced}), with two commonly reported techniques – the dipole approximation method, where both the tip and sample are treated as point dipoles, and the sphere model using Eq. (3) with the tip modeled as a sphere with radius equal to the radius of curvature of the tip (evaluated using Maxwell's stress tensor method, Eq. 3) and the distance between the bottom of the sphere and the sample surface is 5 nm. For the sphere model, the polarizability of the tip, α_t , is calculated using Eq. 2, with $a = 20$ nm and the dielectric constant of gold at 785 nm, $\epsilon = -21 + 1.68i$, while the electric and magnetic field components were calculated at a distance of 5 nm above the surface of the rod using FDTD simulations. Figure 3b shows the line profiles of the out-of-plane (z-component) of the photo-induced force along the middle of the long axis of the nanorod, calculated using the three different evaluation methods. The dipole approximation method underestimates the magnitude of the forces by a factor of ~ 30 , compared with the other two methods, because the tip is

compressed into a point (with radius = 1 nm) that is placed 5 nm away from the rod. Since this method ignores the geometry of the tip, the width of the photo-induced force enhancement peaks, induced by the strong near-fields at the edges of the plasmonic nanorod, and their decay away from the plasmonic structure are underestimated. A better agreement with our model of a realistic tip is achieved when we model the tip as a finite sphere. In this case, the magnitude of the force is on the same order, but both, the magnitude and the width of the peaks are still underestimated by $\sim 20\%$. Therefore, in the following we use the Maxwell's stress tensor method and a realistic tip geometry for the calculation of $F_{induced}$.

Figure 3c, which shows the 2D map of F_z calculated for the gold nanorod, reveals the symmetric hot spots at the tips of the rod. In comparison, the experimental force map also reveals the strong hotspots at the ends of the rods and a diminished amplitude at the center of the rod. Note that in both the experiment and calculation, the strongest force enhancements occur along the middle of the edges of the rods, rather than at the apexes. This is due to the radius of curvature of the tip being close to the width of the rod (25 nm) resulting in a smearing of the distribution of the optical force intensity at the edges. Thus, using a tip with a smaller radius of curvature will result in better resolution of the features around sharp edges.

To understand the vectorial contributions of the near-field photo-induced forces to the signals detected in experiments, we calculate the z-component (F_z , out-of-plane) and the x-component (F_x , along the long axis of the rod) of the induced force along the middle of the rod as shown in Figure 3d. It can be seen that the amplitude of F_x is ~ 5 times weaker in comparison to F_z and that their line shapes are significantly different. Thus, we assume that most of the experimental signal comes from the out-of-plane component of the optical force, as expected for photo-induced force detection using AFM tapping mode^{41,42} (for simplicity, we only consider the

contribution of the x-component of the in-plane forces). The contribution of F_y to the overall force is weaker, or at best comparable to the x-component, depending on the spatial location where the forces are evaluated, as shown in Figure S4). In Figure 3f, we compare the calculated line profile of F_z , with the experimental photo-induced force signal along the long axis of the rod (as shown in Figure 3e), both normalized to their maxima. The maximal calculated enhancement of F_z at the edges of the rod, compared with the center of the rod, is ~ 10 -fold, which is in good agreement with experiments (Figure 3f). Additionally, the calculated peak shape and position at the right edge of the rod, are in good agreement with the experiment. The width of the peak in calculations is slightly overestimated compared to the experiment, most likely due to the radius of the tip being smaller than what was used in the calculations. However, the agreement is poorer for the left edge of the rod, where the measured peak position is shifted significantly from predictions, and the shape of the peak is characterized by a smaller shoulder, close to the center of the rod. The reason for this disagreement is most likely because the surface of the rod imaged in the experiment, is not smooth. As seen in the topography map (right panel of Figure 3e) and in the corresponding height profile (blue line, Figure 3f), a small parasitic feature appears towards the left of the center of the rod, resulting in a kink in the height profile. This kink is also present in the force image (left panel of Figure 3e) and in the line profile of the force intensity (Figure 3f), and distorts the symmetry of the plasmon resonance in the structure.

Moving to a more complex system, we also analyzed gold disk dimers consisting of 150 nm diameter disks separated by a 30 nm gap. In Figure 4 we show the experimental map of the photo-induced force intensity for a dimer structure, with all experimental parameters similar to the gold nanorods case. We also compare the line profiles of the calculated F_z for this structure with the experimental signals along the long (x-axis) and the short (y-axis) axes of the dimer.

From Figure 4a, we see that the calculated optical force profile through the gap of the short axis of the dimer agrees well with the experimental force intensity profile. Both the calculation (8x) and experiment (5.5x) show a ~ 7 -fold enhancement in the optical force at the gap, compared to a spot on the substrate 150 nm away from the gap. However, the agreement between calculation and experiment is not as good along the long axis (Figure 4b). While the peaks due to force enhancements at the edges and in the gap are both present in the calculation and experiment, the position and width of the peaks are considerably different and asymmetric in experiments. This discrepancy is most likely due to a misalignment of the focused beam spot on the glass substrate (focusing point) and the position of the tip with respect to the center of the beam waist. We calculate the F_z line profile for a misalignment of the focusing point with respect to the AFM tip by 0, 200 and 400 nm (Figure 4c). It can be observed that moving the center of the beam waist away from the central axis of the tip, even by a few hundreds of nanometers, breaks the symmetry of the structure, leading to a reduction in the amplitude and deviations of the peak positions of F_z at the gap and the edges of the dimer. These results highlight the importance of careful tip alignment with respect to the center of a tightly focused beam spot for tip-based optical force measurements, in particular for deterministic analysis of symmetric plasmonic structures.

Using this technique, arrays of plasmonic nanostructures can be imaged over few 10s of microns. This capability is demonstrated in Figure 5a, where we mapped an array of plasmonic gold dimers with the incident laser polarization parallel to the long axis of the dimers. Figure 5b shows the top-center dimer of the array (same as in Figure 2b). This example shows the usefulness of photo-induced force microscopy to study inhomogeneities of optical near-fields (Figure 5b), as well as the heterogeneity in local field distributions for nominally equal

nanostructures fabricated by a precision tool such as e-beam lithography. As such, photo-induced force microscopy will be a welcome addition to the toolbox of nanoscale characterization methods, and will find useful applications for the characterization of precisely manufactured nanostructures, surface-enhanced Raman scattering (SERS) substrates and photocatalysis.

In conclusion, photo-induced force microscopy can simultaneously record the topography and the optical near-fields of a sample. We observed that the measured force intensity enhancements in gold disk dimers and nanorods agree well with Maxwell-Stress tensor simulations when a realistic tip-sample geometry is modeled, and can be used to estimate polarization-dependent electric field enhancements. Finally, we believe that photo-induced force microscopy is a promising technique to characterize heterogeneities of precisely manufactured nanostructures, of SERS substrates and heterogeneous (photo-)catalytic samples.

Supporting Information Available: Details of the fabrication process, spatial mapping of the incident beam spot, the calculated optical response of the gold dimer and nanorod structures, and the evaluation of the in-plane and out-of-plane components of the optical force, are included in the Supporting Information. This material is available free of charge via the Internet at <http://pubs.acs.org/>.

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Author Contributions

These authors contributed equally to this work.

Notes

The authors declare no competing financial interest.

Figure 1. (a) Schematic of the setup used for photo-induced force imaging of gold nanorods and dimers. The inset is a cartoon indicating the dipole-dipole interactions between the photo-excited sample and the AFM tip. Scanning electron microscope images of (b) gold disk dimers (c) gold nanorods, fabricated on a glass cover slip.

Figure 2. (a) FDTD simulations of the electric field ($|E|$) distribution on gold disk dimers (for disk radius = 150 nm, gap size = 30 nm and height = 35 nm), calculated for longitudinal (top panel) and transverse (bottom panel) polarizations, at the wavelength $\lambda = 785$ nm. (b) Measured photo-induced force intensity maps on the fabricated gold disk dimers, with longitudinal (top panel) and transverse (bottom panel) polarizations at $\lambda = 785$ nm. The white arrows indicate the direction of the in-plane polarizations. (c) Representative AFM topography image of the same gold disk dimer shown in the bottom panel of Fig. 2b, along with the height profile measured along the axis of the dimer (gray line).

Figure 3. (a) The out-of-plane (z -) component of the optical force ($F_{induced}$) near a gold nanorod (length = 110 nm, width = 25 nm and height = 35 nm), calculated as a function of the integration box height h (using Eq. 3). The inset is a schematic showing the geometry with the integration box used for calculations. (b) Right axis: F_z calculated using the dipole approximation method (blue trace). Left axis: F_z calculated using the Maxwell's stress tensor method (MST) for the case when the tip is modeled as a sphere with radius = 20 nm (dotted black line) and for the case of a realistic tip with radius of curvature = 20 nm ($F_{induced}$) (solid black line). (c) 2-D map of F_z around the gold nanorod (left) and a zoomed-in image of the area enclosed by the green rectangle (right), calculated using MST. (d) Line profiles of $|F_z|$ and $|F_x|$, plotted along the center of the long axis of the rod, calculated using MST. (e) The measured photo-induced force intensity map (left) and the corresponding topography image (right) of a gold

nanorod fabricated on a glass substrate. (f) Right axis: Height profile of the rod along the line shown in the right block of Fig. 3e. Left axis: Normalized force intensity profile measured along the white line shown in the left block of Fig. 3e (solid black line) and the calculated line profile of F_z along the center of the long axis of the rod.

Figure 4. Line profiles of $|F_z|$ (calculated out-of-plane component of the optical force) and the measured force intensity on a gold disk dimer structure along (a) the short (y-) axis and (b) the long (x-) axis of the structure. The insets show the measured photo-induced force intensity maps on the dimer. The white lines represent the region used for plotting the line profiles. (c) Line profiles of $|F_z|$, calculated for three different positions of the center of the beam waist ($1/e^2$ width = 800 nm) with respect to the central axis of the tip. The three positions are at the center of the dimer gap (black line), 200 nm away from the center (red line) and 400 nm away from the center (green line). For all calculations, the diameter of each disk in the dimer was $d = 150$ nm, thickness = 35 nm and a dimer gap size = 30 nm.

Figure 5. (a) Photo-induced force image mapped over an array of plasmonic gold dimers. The heterogeneity of near-field enhancements in precisely e-beam fabricated plasmonic dimers of nominally identical geometry can be clearly observed - a capability of widespread interest for precise nanomanufacturing, SERS and photocatalysis applications. (b) A zoom-in of the plasmonic dimer in the top-center of the array (a), with the incident laser polarization parallel to the long axis of the dimers. Here, field-inhomogeneties of the individual dimer can be clearly imaged.

Table of Content figure:

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