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Grating-Coupling of Surface Plasmons onto Metallic Tips: A Nanoconfined Light Source

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ABSTRACT

We describe and demonstrate a new nanometer-scale broadband light source. It is based on the grating-coupled excitation of surface plasmon polaritons (SPPs) on the shaft of a sharp conical metal taper with a tip radius of few tens of nanometers. Far-field excitation of linear nanoslit gratings results in the resonant generation of SPPs traveling over more than 10 μm to the tip apex and converging to an intense radiative local light spot. Such nanofabricated tips are expected to find various applications in nanospectroscopy, overcoming problems with background illumination in apertureless microscopy.

The control of light propagation and localization on a subwavelength scale is one of the key challenges in nanophotonics. In this respect, surface plasmon polaritons (SPPs)[1] in metallic nanostructures[2] are especially versatile optical excitations. On the one hand, they promote surface-bound electromagnetic transport on nanometer[3,4] or micrometer[5−9] length scales. On the other hand, localized surface plasmons[10] and the associated field enhancements can lead to the spatially selective generation of nonlinear optical signals.[11−15] Because of these superior properties, surface plasmons are expected to bridge the gap between electronics and photonics in integrated systems.[16]

An efficient transformation of traveling delocalized SPPs into highly localized excitations is therefore of central importance to achieve bright illumination of confined volumes. This will be crucial in various applications ranging from optical-electronic interconnects to near-field optical microscopy and nanosensing.[17] Tapered structures such as wedges[18−20] or cones[21−27] have been proposed theoretically to allow for the concentration of surface plasmons at the apex of these convergent geometries.[28] For various geometries, suggestions have been made to launch surface plasmons onto a sharp metallic tip, e.g., by prism coupling[29] or through a thin metallic coating on a fiber taper[26] near the cutoff radius.[30] Very recent experimental work showed an SPP-assisted enhancement of upconversion luminescence at the end of a two-dimensional triangular structure.[31] While the field enhancement in the taper region was clearly demonstrated, radiative losses and SPP reflection at the edges of such planar films[6,31] limit the efficiency of the vertex excitation. Moreover, three-dimensionally tapered cones promise greater field localization and enhancement than planar structures and can be used in near-field scanning optical microscopy applications.

For a number of apertureless near-field imaging and spectroscopy[30,32] techniques, achieving highly localized light at the apex of a three-dimensional metal taper is very desirable, as this could significantly suppress interferences with a background from far-field illumination. Such background interferences currently affect many experiments in apertureless microscopy, and modulation techniques are often necessary to extract the near-field signal.[33] Other approaches make use of specifically designed probes combining the low background of a small aperture with the localization of a metallic apex.[34,35]

Here, we introduce a novel local light source by spatially focusing SPPs on nanofabricated, three-dimensionally tapered conical tips with apex radii of a few tens of nanometers. One-dimensional gratings are written onto the tip shaft by focused ion beam sputtering, more than 10 μm away from the apex. Illumination of the grating with a broadband femtosecond laser leads to resonant excitation of SPPs, which

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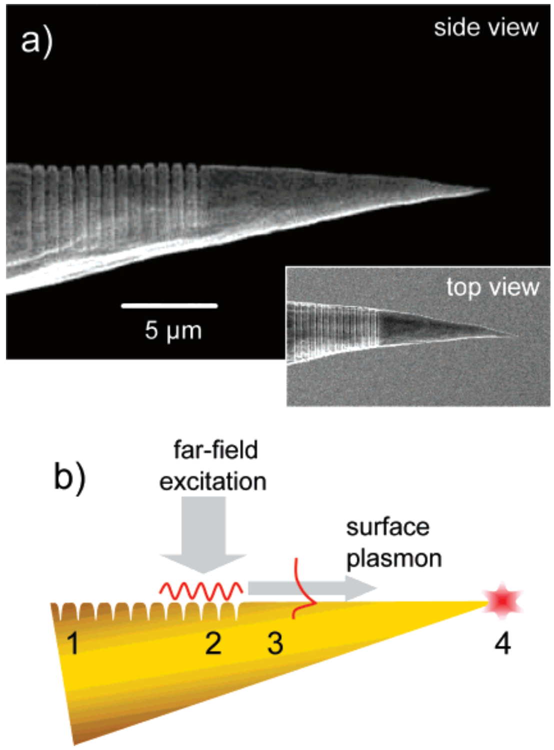

Figure 1. (a) Scanning electron microscope images of a conical metallic tip with a grating coupler on the shaft prepared by focused ion beam sputtering. (b) Principle of the nonlocal excitation of the tip apex. Far-field radiation excites surface plasmon polaritons on the grating, which propagate along the shaft toward the tip apex, where they are reradiated into the far field.

travel to the tip apex and are reradiated. This long SPP propagation length allows us to realize a new local light source, carrying potential for a significant reduction of background illumination in apertureless imaging and spectroscopy applications.

A scanning electron microscope image of one of the nanofabricated metallic tips is shown in Figure 1a. It is based on an electrochemically etched gold tip[36] with a radius of curvature at the tip apex of about 20 nm and an opening angle of 15°. SPPs can be excited in periodic structures on metallic surfaces via grating coupling.[1,37] We make use of this effect by writing a linear grating with a periodicity of approximately 750 nm onto the tip shaft by focused gallium ion beam sputtering, several micrometers away from the tip end. Such gratings have been prepared on a number of tips with different tip shapes and distances from the tip end.

The tip is illuminated with light from a titanium:sapphire laser oscillator, either in continuous-wave operation or mode-locking and generating broadband 7 fs optical pulses at a center wavelength of 800 nm (Femtolasers Inc., Rainbow).[38] The light is focused onto the tip shaft to a spot size of about 5 μm at close to normal incidence (Figure 1b). For focusing, a 20× microscope objective with a numerical aperture of 0.35 and a working distance of 20.5 mm is used. The light scattered from the tip out of the figure plane is collected with a second objective and imaged onto either a video camera or the entrance slit of a spectrometer. The illuminating microscope objective is mounted on a piezoscanner, allowing for a precise positioning of the optical focus on the tip shaft.

Optical microscope images of the light scattered from the tip are recorded for various focus positions of the illuminating light. In Figure 1b, different illumination positions are labeled by the numbers 1 to 4, referring to illumination on the grating on the far side from the tip apex (1), on the grating near its right border (2), between the grating and the tip apex (3), and on the tip apex (4).

At normal incidence on the grating, the excitation of SPPs is expected for wavelengths of the incident light that are close to the grating period.[1,37,39] Therefore, a grating period of 750 nm was chosen to allow for efficient SPP generation with the titanium:sapphire laser. The scattered light images for these four illumination positions and monochromatic illumination at 765 nm are shown in the image series of Figure 2. For illumination on the far left side of the grating, strong scattered light is observed from the grating itself (1). Moving the focus to the right side of the grating, one observes an intense signal from the tip end (2). This image shows clearly that SPPs are very efficiently excited at the grating, propagate over a distance of more than 10 μm without strong scattering losses, and are reradiated into the far field at the apex of the tip. It represents an experimental verification of the concept illustrated in Figure 1b. From a comparison of the signal levels scattered off of the grating with those from the tip end, we estimate that, for optimized coupling conditions, the total power of the light scattered from the tip apex amounts to 0.1−1% of the incident light, i.e., about 1−10 μW for an incident power of 1 mW.

This intense radiation from the tip apex is only observed for a polarization of the incident light perpendicular to the grooves, i.e., for the polarization in which SPP excitation is possible.[1] The comparatively weak scattering from the region between the grating and the tip end indicates that only minor scattering losses are present on the smooth part of the tip shaft. We have observed this strong nonlocal tip excitation for several of our nanofabricated tips, and the best results were obtained for tips with minimal surface roughness between the grating and the tip apex.

The optimum choice of the distance between the grating and the tip apex poses a trade-off between the propagation losses that increase with this distance and the desire for an excitation region well separated from the apex to suppress the direct far-field illumination of the apex. We have found distances of 7 μm or larger (15 μm for the data shown) to be sufficient in order to clearly separate the grating excitation from the apex. The results for two further tips are presented in the Supporting Information. One of those examples illustrates that the nonlocal apex excitation can be frustrated by SPP scattering losses resulting from the presence of a surface defect between the grating and the tip apex.

When the illumination is moved toward the tip apex and off the grating (3), the localized light spot at the tip apex disappears due to a lack of efficient SPP excitation on the unstructured shaft. In image 4, the tip apex is directly illuminated, and scattered light is found from a less-confined region close to the tip end. Moving the illumination focus

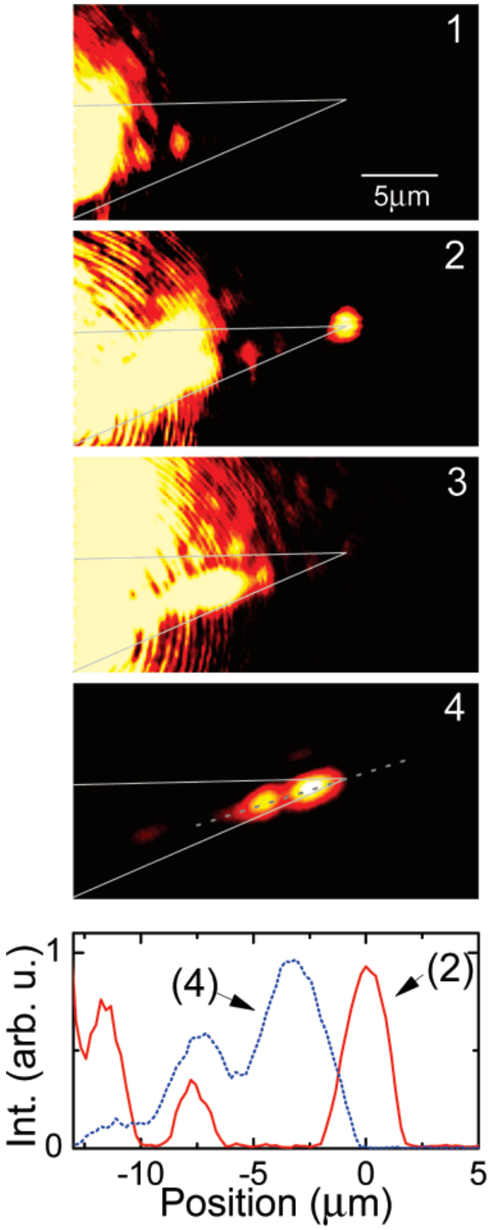

Figure 2. Series of microscope images recorded for illumination of the tip at the four positions indicated in Figure 1b. Image 2 demonstrates the efficient nonlocal excitation of the tip apex via illumination of the grating. The graph below the image series displays sections through Images 2 and 4 corresponding to the dashed line in Image 4.

in image 4 further to the right and eventually away from the tip does not shift the scattering pattern, but only results in a decrease of the scattered intensity. The Supporting Information contains a movie file with an entire scan of the laser focus along the tip.

It is important to note that the maximum intensity of the scattered light in image 4 is not at the very end of the tip, in stark contrast with image 2. The graph below the image series in Figure 2 displays sections of the images 2 (solid red) and 4 (dotted blue) along the tip axis, as indicated by the dashed line in image 4. For illumination with far-field light, the scattering cross section depends on the illuminated metallic area on the tip shaft and on the local field enhancement. Because of the reduction in tip diameter toward the apex, the intensity scattered from the apex itself becomes small compared with the somewhat thicker region close to the tip end. We have made very similar observations in recent far-field measurements involving the generation of nonlinear signals (second-harmonic and multiphoton electron emission) at the apex of such metallic tips.[11] These experiments showed that the nonlinear signals were generated at the tip apex in a region where the linear (elastic) scattering from direct illumination was, as in this work, substantially reduced. In that study, light localization to a region of few tens of nanometers in diameter at the tip apex was unambiguously identified in subsequent near-field measurements.

The spatial shift between the scattering maxima in images 2 and 4 and its correspondence with previous nonlinear experiments has two major consequences. First, it clarifies that the emission in image 2 is indeed stemming from an area much smaller than the optical focus. Second, it demonstrates the physical difference in the excitation conditions underlying the images 2 and 4. Whereas image 4 is a result of far-field excitation and scattering, evanescent surface waves propagating along the tip shaft are responsible for the scattered light from the tip apex in image 2.

The SPPs excited in the grating travel over a distance of more than 10 μm all the way to the tip end, where they are reradiated into the far-field. Radiation damping is suppressed for the regions on the tip shaft, as long as the circular cross section of the conical tip varies slowly over a distance of the SPP wavelength.[21] This condition fails at the very end of the tip, resulting in strong SPP scattering, predominantly into the far-field, as experimentally shown, but also back onto the tip shaft. For the same kind of gold tips as those used in the present experiments, we have measured large local field enhancements of about 10.[11,12] Because of these large field enhancements, the radiation damping of SPPs at the tip apex is very effective, the damping rate being proportional to the square of the local field strength.[21,23,24] As a result of this efficient spatial excitation transfer, the size of the excitation spot is reduced from a few micrometers in and near the grating to only few tens of nanometers. This "superfocusing" has previously been predicted theoretically[21,23,24] and is experimentally demonstrated in image 2 in the regime of linear optics.

Equally strong light localization at the apex of sharp metal tips is usually only achieved by making use of the field-enhanced optical nonlinearities of these tips, for example, by locally generating second-harmonic light.[11−15] In using such frequency-converting nonlinear techniques for near-field microscopy, substantial limitations arise from the required intense direct illumination of the tip−sample geometry with the fundamental laser beam and from the much smaller conversion efficiency than in the presented linear nonlocal apex excitation scheme.

In the following, the resonant nature of this nonlocal excitation of the tip apex is discussed. In our experiments, the optimum condition for the tip excitation was found for slightly defocused illumination of a series of slits in the grating, thus resonantly enhancing the coupling efficiency

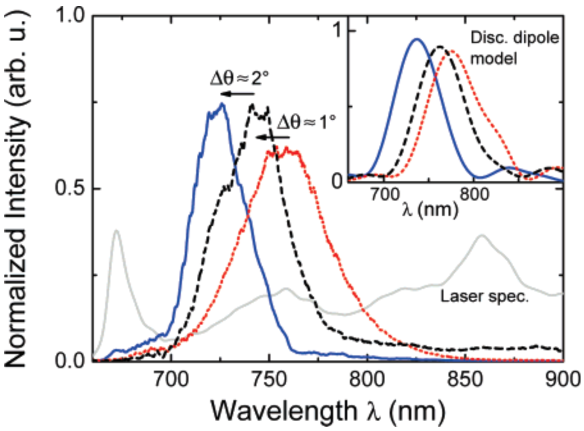

Figure 3. Recorded scattering spectra from the tip apex for nonlocal excitation at the grating near normal incidence ($\theta \sim 0°$, dotted, red) and for two slightly larger angles of incidence, incrementally increasing by about one ($\Delta\theta \approx 1°$, dashed, black) and two degrees ($\Delta\theta \approx 2°$, solid, blue). The spectra are normalized to the broadband input laser spectrum (solid, gray). A shifting and broadening of the spectra is observed for a variation of the angle of incidence. A similar behavior is found within a phenomenological discrete dipole model (inset) for angles of incidence of 3.5° (dotted, red), 4.5° (dashed, black) and 6.5° (solid, blue).

to SPPs.[40] Depending on the angle of incidence, different SPP frequencies ω can be excited, as is well-known from the SPP dispersion relation[1] at a planar metal−vacuum interface $\omega(k_{SP}) = c|k_{SP}|((\epsilon_m + 1)/\epsilon_m)^{1/2}$. Here, ϵ_m is the dielectric constant of the metal, k_{SP} is the surface plasmon wave number, and c the vacuum speed of light. We have experimentally verified this behavior by spectrally resolving the light scattered from the tip apex under illumination of the grating with the broadband 7 fs optical pulses from the laser oscillator. In Figure 3, three scattering spectra are displayed for different representative angles of incidence near 0° degrees (normal incidence) and with a difference of about 1° and 2° between them, respectively. The absolute angle of incidence could not be exactly determined in the experiment due to a lack of a well-defined specular reflection from the tip shaft. The spectra are normalized with respect to the laser spectrum, which is shown as the solid gray line. One observes clear resonant features at wavelengths of 720, 740, and 760 nm, close to the grating period. Resonances in this wavelength range are expected from the SPP dispersion relation on the planar interface, energy conservation $\omega_L = \omega(k_{SP})$ and momentum conservation $k_{SP} = k_{\|} + p \cdot 2\pi/a_0$. Here, ω_L denotes the frequency of the incident light with in-plane vector component $k_{\|}$, a_0 the grating period, and p the diffraction order (here, $p = \pm 1$).[1,37] The strong dependence on the excitation conditions evidences that this tunable resonant behavior is indeed an effect of the collective excitation of the grooves and not a pure resonance of the tip itself.[36]

We have performed model calculations for a phenomenological scalar discrete dipole model including SPP scattering[41] in which the grooves of the grating and the tip apex are simulated as point dipoles on a line. The incident fields are assumed to impinge only on the grating, and the resulting field at the apex is self-consistently calculated as a function of wavelength and angle of incidence. The inset in Figure 3 shows the results of these calculations for angles of incidence of 3.5° (dotted, red), 4.5° (dashed, black), and 6.5° (solid, blue), leaving the polarizability of the grooves as a free parameter. The qualitative agreement between these model calculations and the experimental results supports the interpretation of the grating resonance effect. The single groove polarizability in the simulations was chosen such that up to 10% of the incident radiation is converted to SPPs, a value comparable to those recently calculated for linear nanoslit apertures in a metallic film.[42] Nonetheless, a parameter-free quantitative calculation, taking into account the correct three-dimensional geometry and the grating, is certainly desirable. In this context, the recent numerical approach from ref 27, which has successfully quantified absorption, scattering, and reflection on a conical taper, could perhaps be adapted to the present geometry.

Interesting questions arise concerning the dynamics of this new local light source. SPP resonances on such gratings are radiatively broadened,[43] with SPP lifetimes ranging from a few tens to a few hundreds of femtoseconds.[39] In our case, the resonance width is mainly determined by radiative coupling and by the number of grooves present and illuminated on the shaft of the tip. Furthermore, the particular shape and depth of the grooves may influence the resonant excitation. The experimentally observed line widths suggest the possibility of launching SPP wavepackets with a duration on the order of 10 fs onto the tip shaft. Theoretical results indicate that significant chirp may be acquired by an SPP wavepacket upon propagation on a tapered conical waveguide.[21,23,24] In this case, dispersion compensation of the incident pulse will have to be applied to achieve the shortest possible pulses at the tip end. Further experimental work will be necessary to resolve this issue, e.g., by investigating optical nonlinearities at the tip apex[11−15] and/or by using adaptive control schemes.[44] Together with near-field optical experiments, this will also help to quantify the absolute local intensities at the tip apex and the achievable field enhancements in such a three-dimensionally tapered waveguide.[21]

In conclusion, we have demonstrated an efficient nonlocal optical excitation of the apex of a nanostructured metal taper by using grating-coupling of surface plasmon polaritons. These surface plasmon polaritons propagate over more than 10 μm along the tip shaft toward the apex of the tip, where they are reradiated into the far field. The tip emission is spectrally tunable by varying the angle of incidence of the illumination. Although we have not yet performed near-field optical experiments to directly characterize the field distribution at the tip apex, the presented results indicate that such nanofabricated tips will serve as bright nanoscale light sources, for example, in apertureless microscopy and spectroscopy, where they could lead to a significant reduction of the direct illumination of a sample in proximity to the tip apex.[45] It will be very interesting to explore and exploit the ultrafast temporal characteristics and the near-field localization of these tips.

Supporting Information Available: Scattering images recorded from two additional nanofabricated tips (PDF).

Movie file showing an entire spatial scan of the laser illumination along the tip (AVI). This material is available free of charge via the Internet at http://pubs.acs.org.

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