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Review Paper

Light Confinement at Ultrasharp Metallic Tips

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In this article, two alternative experimental strategies for reaching light confinement at metallic nanotips are discussed. The first approach utilizes optical field enhancement to localize nonlinear optical signals at the tip end. The second approach employs surface plasmon polaritons propagating coherently along the tip shaft and converging at its apex.
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1. Introduction

The measurement and manipulation of light confinement on the nanoscale are among the primary objectives in nano-optics. The exceptional optical properties of metals can be utilized to create nanostructures with strongly enhanced local fields. In the case of sharp metallic needles, these field enhancements, being the optical counterpart of the electric lightning rod effect, naturally occur at the apex of such structures. This property has led to the development of apertureless near-field optical microscopy with spatial resolutions on the order of 10 nm.[1–3] Unfortunately, this method is sometimes complicated by a large background and interferences between the wanted signal originating from the tip apex and unwanted light scattering from the tip shaft. Owing to the resulting drastic reduction of signal-to-noise ratios and — in the worst case — loss of spatial resolution, modulation techniques are often necessary to extract the near-field signal.[4] Significant background contributions are particularly detrimental in elastic, i.e., frequency conserving, experiments such as local absorption spectroscopy.[5] Thus, an optimized light localization to the tip apex and a background suppression inherent to the near-field probe are highly desired in many near-field microscopy applications.

In this article, we discuss two alternative, recently investigated approaches towards improved light confinement at ultrasharp metallic tips. The first method utilizes of the localization of nonlinear optical signals in the presence of field enhancement.[6] In the second part, a newly developed scheme for realizing a nonlocal apex excitation via surface plasmons traveling on the tip shaft is presented.[7]

2. Nonlinear Light Localization

In the regime of nonlinear optics, the background problems of linear experiments can disappear almost entirely, and the properties of such metallic tips may be completely dominated by field enhancement at the very end of the tip. We begin by looking at the scattering of light from a sharp gold tip, which was electrochemically etched to a radius of curvature of about 20 nm. Illumination of such a sharp tip with femtosecond light pulses results in nonlinear frequency conversion.[8–11] We detect this optical frequency conversion in the experimental configuration shown in Fig. 1(a). A given tip is illuminated with 7-fs light pulses from a Ti:sapphire oscillator, linearly polarized along the tip axis and focused down to a spot size of 1.5 μm by a reflective Cassegrain microscope objective (0.4 numerical aperture; 10 mm working distance). Compared with conventional refractive objectives, such a mirror objective has the great advantage of very little spectral dispersion, which is essential for the efficient generation of nonlinear optical signals. The average incident power was limited to about 10 mW to avoid tip damage. The tip is mounted on a piezo scanner, which allows it to be raster scanned through the laser focus in the plane perpendicular to the optical axis. The backscattered light is separated from the incoming laser by a dichroic beam splitter, dispersed in a monochromator and detected using a liquid-nitrogen-cooled charge-coupled-device (CCD) camera, yielding a spectrum at every point in the scanning routine.

An example of a spectrum of the frequency-converted light from a gold tip is shown in Fig. 1(b). The generated light is typically composed of the second-harmonic of the laser (at wavelengths about 450 nm) and a broad continuum in the range between 450 and 700 nm, which very likely stems from two-photon-induced luminescence.[9] The relative amplitudes of the second harmonic and the continuum contributions vary for different tips, which might stem from different microscopic tip structures and/or crystalline orientations at the tip apex.

A small fraction of the fundamental laser light scattered back from the tip and transmitted through the dichroic beam splitter serves as a reference. Figure 2(a) shows this backscattered laser light as a function of the focus position on the tip. The image clearly resembles the tip shape, which is indicated by the white lines. Close to the tip end, where the tip becomes thinner than the laser focus, the scattered light intensity decreases owing to the limited linear scattering cross section. The image shown in Fig. 2(b), displaying the photon rate detected in the wavelength range outside of the laser spectrum, appears markedly different. One observes an intense concentration of this nonlinear light generation at the very end of the tip. This localization of the frequency-converted light is a direct consequence of the enhanced field at the tip apex, and it is only present for a polarization parallel to the tip axis. Moreover, in the case of the second-harmonic contribution, the broken spatial sym-

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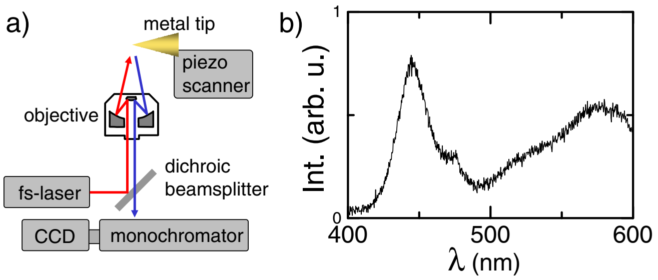

Fig. 1. (Color online) (a) Experimental setup for detection of nonlinear frequency conversion from metallic tips. Optical pulses from a fs-laser are focused with a Cassegrain mirror objective onto the tip. An image is obtained by scanning the tip through the laser focus and by detecting the spectrally dispersed backscattered light. (b) Spectrum of frequency-converted light for one of the tips, consisting of a second-harmonic contribution and a broad continuum.

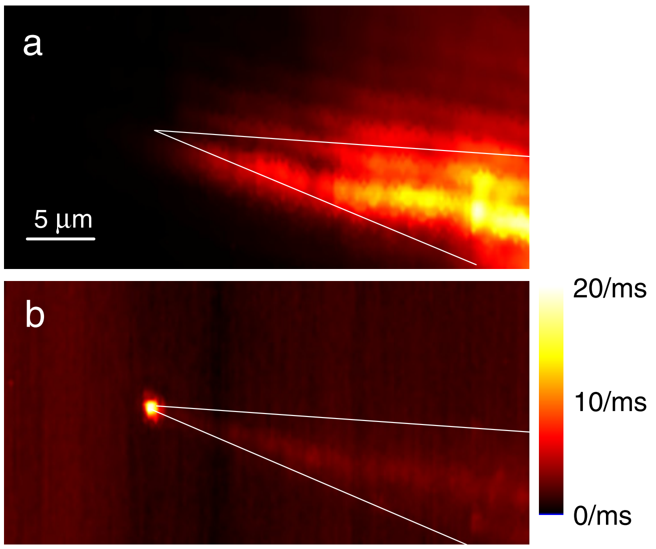

Fig. 2. (Color online) Strong localization of optical frequency conversion at the apex of a gold tip (indicated by white lines). (a) Backscattered fundamental laser light. (b) Frequency-converted light confined to the tip apex. Color bar labeled with detected photon rates.

metry at the apex is important for generating the second-order nonlinear polarization in the otherwise centrosymmetric material gold.[11)]

A comparison between the backscattered nonlinear light from the apex and that of the flat shaft regions allows for a rough estimation of the field enhancement α at the tip, as outlined in the following: The local electric field at the apex $E_{\rm loc}$ is larger than the incident field $E_{\rm inc}$ by a factor of the field enhancement, $E_{\rm loc} = \alpha E_{\rm inc}$. In the case of second-harmonic generation, the total signal strength S scales with the fourth power of the local electric field times the area $A_{\rm tip}$, from which the signal is stemming, i.e., $S_{\rm tip} \propto A_{\rm tip}|E_{\rm loc}| = A_{\rm tip}\alpha^4|E_{\rm inc}|^4$. On the smooth parts of the shaft with no field enhancement, the weak signal is generated in a larger area given roughly by the illuminating spot size $S_{\rm shaft} \propto A_{\rm spot}|E_{\rm inc}|^4$. Thus, the relative signal levels can yield an estimate of the field enhancement,

$$\alpha \approx \left(\frac{A_{\rm spot}\, S_{\rm tip}}{A_{\rm tip}\, S_{\rm shaft}}\right)^{1/4}. \qquad (1)$$

In the measurement shown in Fig. 1, the second-harmonic light from the tip is at least a factor of ten stronger than very weak signals discernible from the shaft, so that a field enhancement $\alpha \approx 15$ is estimated for $A_{\rm tip} = 20 \times 20\,{\rm nm}^2$ and $A_{\rm spot} = 1.5 \times 1.5\,\mu{\rm m}^2$. This value is in good agreement with those from previous experiments[11)] and field enhancements obtained from theoretical calculations for gold tips of similar sharpness.[8)] We have also found comparable field enhancements in a recent study investigating femtosecond free-electron emission from metal tips.[6)] In that process involving an eighth-order power dependence on the local electric field, an even stronger suppression of the nonlinear signal from the shaft was observed.

3. Linear Light Localization

In the previous section, we demonstrated light localization at the tip end on the basis of nonlinearly enhanced backscattering. Yet, the high optical intensities necessary for this process may be undesirable in many circumstances. In particular, these include experiments in which a bright illumination of an investigated sample in close proximity to the tip needs to be avoided. Moreover, a critical issue was not addressed in this study, namely, the striking mode mismatch between the micron-sized illuminating far-field mode and the nanometer-localized "hot spot" at the apex of the tip. Figures 3(a) and 3(b) illustrate this mode mismatch together with a strategy to overcome it. In Fig. 3(a), the direct far-field illumination of a tip is depicted, and most of the incident optical power misses the tip, potentially resulting in unwanted far-field illumination of a sample near the tip. In contrast, Fig. 3(b) shows a scheme in which evanescent surface waves, so-called surface plasmon polaritons (SPPs),[12)] travel on the metallic cone and converge in the apex. Previously, surface plasmon polaritons have been suggested theoretically to facilitate this form of focusing in two-dimensional wedges[13–15)] or three-dimensional cones.[16–22)] Experimentally, a challenge in realizing such a scenario is how to generate surface plasmons on the conical tip. Previous suggestions include prism coupling[12,23)] or excitation through a thin metallic coating on a fiber taper.[21)]

In the present work, SPP excitation by introducing gratings on the shafts of metallic tips is realized, as shown in Fig. 3(b). Grating coupling is known to be an efficient means of a resonant SPP excitation.[12,24)] Using focused ion beam etching, linear gratings with a period of about 750 nm and a depth of 200 nm were written onto the shafts of various tips, at distances of 7 μm or larger. These distances are both sufficiently large to clearly spatially separate the excitation on the grating from the apex, but still sufficiently short to prevent exceeding propagation losses of SPPs between the grating and the tip apex. A side-view scanning electron micrograph of one of the functionalized tips is displayed in Fig. 3(c). This tip has a 15 μm distance between the grating and the apex.

In the experiment, the tips are illuminated with light from a 7-fs Ti:sapphire laser oscillator, as in the previous section. The light pulses are weakly focused onto the tip shaft, roughly perpendicular to the grating [see Fig. 3(b)] with a microscope objective (0.35 numerical aperture, 20.5 mm working distance) to a spot size of roughly 5 μm. A video

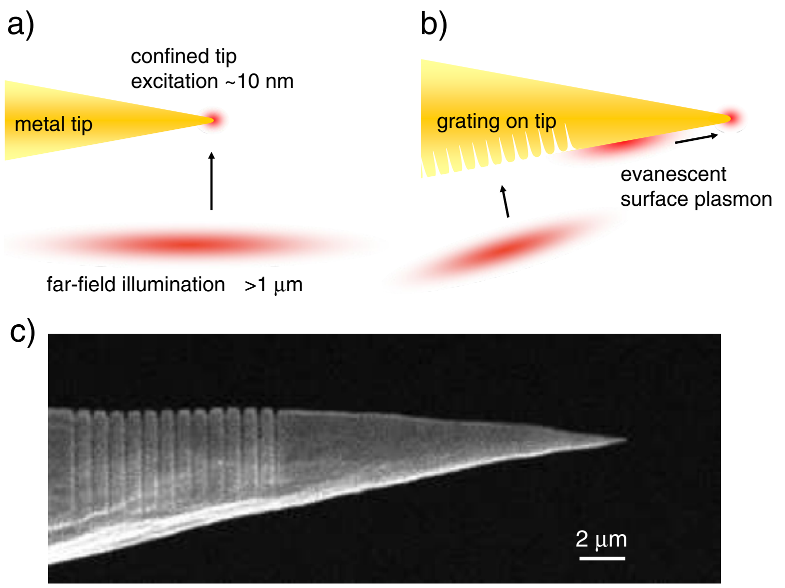

Fig. 3. (Color online) (a) Illustration of the spatial mode mismatch between a micron scale illumination and the nanometer-localized excitation at the tip apex. (b) Scheme for an optimized excitation of the tip apex by surface-bound electromagnetic waves. These surface plasmons are to be excited in the grating on the tip shaft. (c) Scanning electron micrograph of one of the tips prepared with a grating on its shaft.

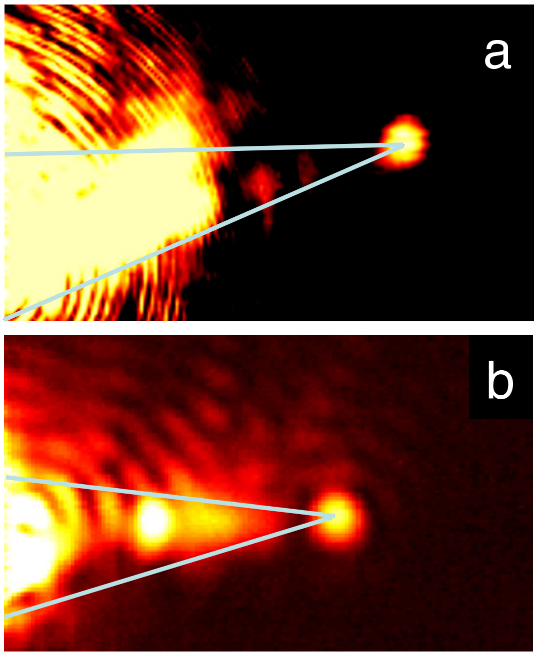

Fig. 4. (Color online) Scattered light images detected from two different tips illuminated at the grating on the shaft. The tip shapes are indicated by the thin gray lines. In both cases, a strong scattering is observed not only from the shaft, but also from the unilluminated tip end. This evidences the nonlocal apex excitation scheme depicted in Fig. 3(b).

camera is used to collect the light scattered off the tip. Figure 4 contains the resulting scattered light images for two different tips, both being illuminated on the grating alone. Besides a strong scattering at the grating itself, an intense light emission from the tip end is found. This directly proves an indirect excitation of the tip end mediated by surface plasmons propagating on the tip shaft from the grating to the apex. Moreover, this interpretation is supported by the fact that the apex emission is only observed for an incident light polarization perpendicular to the grooves in the grating. Only for this polarization, efficient SPP excitation in the grating is expected.[12)]

On a grating, the SPPs generated by scattering at the individual grooves coherently add up to form a standing surface plasmon wave in the grating.[25)] That is, the momentum mismatch between propagating light and the surface plasmon dispersion relation is bridged by a grating vector.[12,24)] This resonant grating mode leaks out at the edges of the groove pattern onto the smooth part of the shaft. On the shaft, the narrowing of the conical taper leads to an ever-increasing spatial concentration of the surface plasmon amplitude towards the apex. As a result of this efficient spatial excitation transfer, the size of the excitation spot is reduced from about five microns in and near the grating to only few tens of nanometers. This "superfocusing" has previously been predicted theoretically.[16,18,19)]

The resonant nature of this excitation process becomes apparent in a further experiment, in which the light scattered from the tip is detected with a spectrometer, as drawn in Fig. 5(a). The recorded spectrum is normalized to that of the incident broadband laser. A typical spectrum obtained from the apex is plotted in Fig. 5(b) (black), together with a Lorentzian fit (gray). One clearly observes a resonance shape with a center wavelength of 757 nm and a full-width of about 50 nm. Because the line width is a measure of the coupling strength between the far-field and the surface plasmon mode, the resonance profile is affected by a number of different factors, including the number of illuminated grooves on the shaft and the particular groove shape and depth. Further work may lead to an optimization of SPP excitation efficiencies by tailoring these parameters. At the present stage, we estimate that up to 0.1–1% of the power incident on the grating in fact reaches the tip end.

4. Conclusions

We have presented experimental results of two different methods to generate a confined light source at the apex of ultrasharp metallic tips. In the regime of nonlinear optics, confinement naturally arises from the field enhancement at the apex. In order to reach comparable localization in linear optics, a different approach was introduced in which a greatly improved apex excitation is facilitated by evanescent

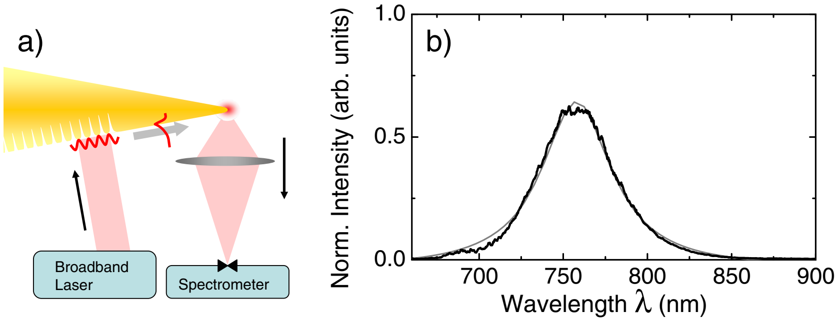

Fig. 5. (Color online) (a) Experimental setup for measuring the spectral response of the functionalized tip. (b) Normalized spectrum (black) of light detected from the apex. A clear resonance behavior is observed. The gray line is a Lorentzian fit to the data.

surface waves. In our opinion, this method of converting traveling SPPs into a localized excitation at the tip end is particularly promising, as it allows for a large spatial separation of the far-field excitation from the apex. Consequently, a significantly reduced background signal in illumination mode apertureless near-field imaging is expected, for example, in transmission geometries. Experiments probing the precise spatial resolution of these sources are currently underway.

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