Cloaked Near-Field Scanning Optical Microscope Tip for Noninvasive Near-Field Imaging

Andrea Alù¹ and Nader Engheta²

¹Department of Electrical and Computer Engineering, The University of Texas at Austin,

1 University Station C0803, Austin, Texas 78712, USA

²Department of Electrical and Systems Engineering, University of Pennsylvania,

200 South 33rd Street, ESE 203 Moore, Philadelphia, Pennsylvania 19104, USA

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Near-field imaging is a well-established technique in biomedical measurements, since closer to the detail of interest it is possible to resolve subwavelength details otherwise unresolved by regular lenses. A near-field scanning optical microscope (NSOM) tip may indeed overcome the resolution limits of far-field optics, but its proximity inherently perturbs the measurement. Here, we apply the recent concept of a "cloaked sensor" to an NSOM device in collection mode, showing theoretically how a proper plasmonic cover applied to an NSOM tip may drastically improve its overall measurement capabilities.

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Near-field scanning optical microscope (NSOM) measurements are currently a well-established technique for resolving the subwavelength details of a complex image [1–3]. As is well known, NSOMs can be operated in two distinct modes: apertureless [2] and aperture [3] mode. In the aperture mode, the tip's opening is used to collect and/ or illuminate the subwavelength detail of objects of interest, whereas in the apertureless operation a very sharp tip is mainly used as a resonant scatterer to enhance and focus light on the subwavelength detail of interest. Recently, apertureless NSOMs have also been used in conjunction with collections of quantum dots localized on the tip to illuminate the detail of interest [4]. Although the apertureless method may ensure a smaller tip size and a larger scattering enhancement for small objects, which results in overall higher resolution, the aperture mode remains more popular, due to its flexibility and simplicity of operation, combined with longer history. In the collection mode, the aperture, few tens of nanometers wide, is used as a stethoscopic probe to capture the small amount of light scattered by the object to be imaged. Apertureless tips require the use of resonant plasmonic materials, which provide large scattering cross sections from the subwavelength tip size, but in the aperture tip a wide range of metallic materials may be used, since the role of the cover surrounding the aperture mainly consists in confining the received signal from the object and guide it towards the optical fiber connected to the tip. Although resolutions up to the order of 20-50 nm may be successfully achieved within this operation, one of the well-known drawbacks of near-field measurements is the inherent disturbance that the proximity of a metallic tip induces on its own measurement [5].

In a series of papers, we have suggested that properly designed plasmonic layers covering passive conducting or dielectric objects may produce a scattering cancellation effect that induces invisibility and cloaking of moderately sized objects [6]. As we have recently highlighted [7], a novel and possibly groundbreaking application of plasmonic cloaking stems from the field penetration inside the cloak, which may be used for noninvasive sensing: a suitably designed plasmonic layer may indeed cloak a sensor for all polarization and incidence angles, without affecting its overall capability to extract a good level of signal and to sense its surrounding. The general concept of "cloaked sensor," introduced [7] for a radio-frequency dipole antenna, may have an impact on a wide range of applications. Here we propose to apply these concepts to NSOM aperture tips in collection mode for near-field optical measurements. It is noticed that the operation of the cloak will inherently reduce the scattering from the NSOM tip, so this technique is not applicable to other NSOM operations, such as in its apertureless functionality, for which maximized scattering is desired. Such operation, however, leads to an inherently strong impact on the fidelity of the measurement. We propose in the following a novel concept for near-field measurements that may combine subwavelength resolution with low noise and high fidelity.

Consider the geometry of Fig. 1, i.e., a spherical aluminum tip of diameter 2a = 150 nm with a circular aperture of diameter 2d = 30 nm at its bottom. The aperture represents the end of a conical aperture carved in metal, which is designed to carry the signal into an optical fiber connected at the back of the spherical tip. The aperture size is associated with the maximum resolution of the instrument. This represents a common model for collection-mode NSOM geometries, widely used in a variety of near-field measurements. Several metals, e.g., aluminum, may be employed in realizing NSOM tips, but in the collection mode the nature of the metal, and, in particular, its scattering properties, are not as crucial as in apertureless NSOM devices, in which scattering enhancement is desirable. The choice of aluminum, therefore, here is arbitrary.

Although the tip represents just the end of a complex imaging apparatus, several thousands of wavelengths large,

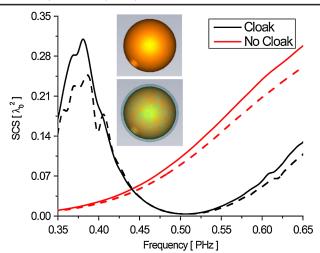


FIG. 1 (color online). Total SCS versus frequency for a typical aluminum NSOM tip (see inset) in collection mode, compared with the same tip covered by a thin plasmonic cloaking layer. Solid lines refer to plane-wave incidence normal to the aperture, dashed lines to plane-wave incidence from the side of the tip, to model different NSOM operations.

the main effect of near-field disturbance on the measurement of a small detail placed few nm away from the aperture is caused by the spherical tip itself. For this reason, in Fig. 1 we report the total scattering cross section (SCS) of the isolated aluminum tip (red, lighter lines; see inset for the geometry). For this calculation, we have used suitable dispersion for the aluminum permittivity reported in the literature [8], including realistic frequency dispersion and losses. It is evident that the aluminum tip, despite its subwavelength size, has a non-negligible SCS at optical wavelengths. In the same figure we report the case of a cloaked tip (black lines), whose shell has been designed by applying the plasmonic cloaking technique [6]. In this case, an optimal shell geometry was found for permittivity $\varepsilon_c \simeq 0.1 \varepsilon_0$ and thickness t = 13 nm. The cloak has been tailored to provide minimum scattering at f = 500 THz, and its permittivity is assumed to have a proper frequency dispersion following a Drude model with $\varepsilon_c = \varepsilon_0 (1 - \omega_p^2 / [\omega(\omega + i\gamma)])$, with $\gamma = 10^{-3} \omega_p$. Effective material properties within this range may be realized at optical frequencies in a variety of metamaterial geometries, like arrangements of nanoparticles [9], mixtures of plasmonic and dielectric materials [10], or specifically tailored nanoimplants [11]. In the figure, we report the total SCS for plane-wave incidence normal to the aperture (as in direct collection and illumination modes; solid lines) and for incidence from its side (as in reflectioncollection operation; dashed lines). It is relevant to stress that the designed cloak is capable of suppressing the scattering from the tip for both polarizations, in both planes and for all incidence angles, for all positions, distances, and forms of excitation and for all spatial wave numbers (even for evanescent waves), despite the asymmetrical shape of the tip. Moreover, the relatively large bandwidth of operation of the plasmonic cloak in Fig. 1, which stems from the

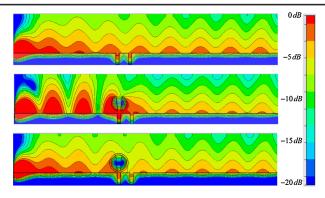


FIG. 2 (color online). Amplitude of the magnetic field on the E plane for a silver surface excited by a molecule at 500 THz (on the far left of each panel) supports a propagating SPP impinging on two narrow slits (top panel). When a tip scans the surface at 20 nm distance (middle panel), inevitable strong disturbance is produced. The use of a plasmonic cloak (bottom panel) suppresses this disturbance, keeping the level of fields induced at the aperture comparable to those without the cloak.

relatively small size of the NSOM tip [12], ensures that these concepts may be practically applied to finite-bandwidth signals. All of these properties, specific to the plasmonic cloaking technique [6], are particularly relevant when one applies these concepts to different modes of operation of practical aperture NSOMs. It should also be mentioned that moderately larger level of losses in the plasmonic shell would not significantly affect the overall cloaking effect, as extensively discussed in Ref. [12]. In principle, the presence of a cloaking layer inherently introduces a finite distance between the probe aperture and the object to be scanned, possibly affecting the resolution of the measurement. However, the inherently thin nature of plasmonic cloaks [6] ensures that its effect in reduction of resolution is little for the range of resolutions typical of aperture-based NSOM measurements.

We report in Ref. [13] the near-field distribution for the bare and cloaked geometries of Fig. 1 for plane-wave incidence impinging normal towards the aperture. The panels confirm how, despite the strong scattering reduction, the plasmonic cloak does not isolate the tip from the surrounding environment, allowing penetration of the fields inside the tip at a level comparable with that of the bare scenario, and ensuring that its capability to collect external signals is not essentially affected.

As a first practical example, consider in Fig. 2 an NSOM tip in collection mode scanning a silver surface supporting a surface plasmon polariton (SPP) wave. The surface is excited by an emitting molecule on the far left of each panel, and is carved with two narrow slits, each 30 nm wide and separated 120 nm from each other. We consider here a realistic permittivity model for the silver material, including frequency dispersion and losses [14]. In the top panel of Fig. 2, we report the SPP distribution (amplitude of the magnetic field normal to the figure) without the presence of the tip, which shows, as expected, a small perturbation of the uniform SPP mode caused by the presence of the two

slits on the silver surface. In the middle panel, we show the full-wave simulation of the same setup scanned by the NSOM tip of Fig. 1, placed 20 nm above the surface. Its presence strongly perturbs the original SPP distribution, creating a strong standing wave between the source (e.g., emitting molecule) and the tip, and drastically modifying the near field of the aperture. The image taken by such NSOM tip in the collection mode will necessarily present some unwanted artifacts due to its own disturbance on the measurement. In the bottom panel, the tip is covered by a proper cloaking material, as in Fig. 1. It is seen that in this case the SPP distribution is restored exactly as if the tip were not there, still allowing a comparable level of field penetration in the aperture. This is achieved despite the fact that the tip is kept at the same distance from the silver surface, and therefore the distance of the outer surface of the cloak from the slits is only 7 nm in these simulations. It is evident how the sensing mechanism may be greatly improved by the presence of this cloaking layer.

In Fig. 3, we report the simulated images at the top end of the NSOM tip in collection mode, proportional to the signal transmitted into the optical fiber connected to the tip in a realistic near-field scenario. We compare the magnitude of the magnetic field at the end of the tip in the three cases of (a) an "ideal" mathematical tip (dotted line) that would pick up the field across the sample, scanning the surface at 20 nm distance with a finite step of 5 nm in the transverse direction; (b) the real bare tip, as in Fig. 1 (red line); (c) the cloaked tip (black solid line). The horizontal axis reports the distance from the source (e.g., emitting molecule) in Fig. 3. The slits are placed at 1 μ m and 1.12 μ m from the molecule, as highlighted in the figure. As in a realistic scanning scenario, the tip moves parallel to the surface, recording the magnetic field amplitude at the tip end. It is seen how the cloaked tip consistently resembles the ideal scenario, without artifacts, whereas the realistic tip scenario creates substantial differences in

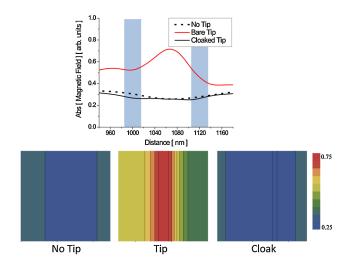


FIG. 3 (color online). Comparison of the sampled images scanned in the setup of Fig. 2 by an ideal mathematical probe (no tip), the bare tip and the cloaked tip of Fig. 1.

the level of the detected fields and its overall distribution, associated with the mutual coupling between the tip and the two slits. For additional clarity, the bottom panels in Fig. 3 report the simulated 2D images in the three cases, showing the significant artifacts generated by the presence of the tip [5], but the proper use of a cloak may drastically reduce their impact.

As another example, consider the geometry in the top panel of Fig. 4, consisting of the same NSOM tip in collection mode, but now imaging the near-field properties of a silver nanoantenna, designed following our previous works [15] to operate around 500 THz. The total length of the nanodipole is 65 nm, with a center-symmetrical gap of 5 nm, used for feeding and tuning purposes [15], and a diameter of 14 nm. These dimensions ensure that the nanodipole may be matched to an optical source (e.g., an emitting molecule, an incoming optical waveguide, etc.) and support its dominant resonance at 500 THz. Notice that at these frequencies, due to the plasmonic properties of the nanodipole, its overall size is extremely subwavelength, and even a narrow tip looks gigantic in comparison (drawing is in scale in Fig. 4).

In the middle left panel of Fig. 4, we show the simulation results for the amplitude of the magnetic field distribution for the emitting nanoantenna at its resonant frequency, with the bare tip at a distance of 22 nm. The dipolar near-zone fields of the nanoantenna are greatly perturbed by the presence of the tip, which collects and senses some of the radiated near field, but at the price of strongly perturbing its pattern, and slightly detuning its resonance (the calculated resonance frequency is shifted in this scenario by about

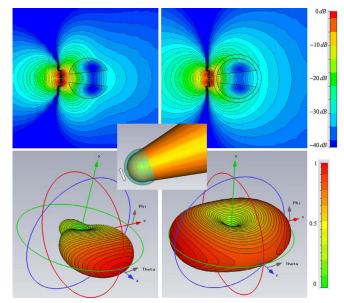


FIG. 4 (color online). Cloaked NSOM tip for nanoantenna measurements. Comparison of the near- (middle panel, magnetic field) and far- (bottom panel) field distributions between cloaked and bare tip measurements, sensing a resonant nanodipole antenna fed by an optical dipole source (e.g., an emitting molecule) at its gap. The geometry details are reported in the text.

 $1\% \simeq 5$ THz). In contrast, the middle right panel presents the same setup, but cloaked as in Fig. 1. The cloak is capable of restoring the dipolar fields of the nanodipole and the resonance properties of the nanoantenna, yet sensing and imaging the unperturbed resonant field distribution. The measured level of fields induced at the aperture is comparable in both cases. Also in this different operation, the cloak provides an excellent mechanism to improve the sensing and scanning operation of the NSOM, ensuring its robustness to different forms of excitations and measurements. In the bottom row of Fig. 4, we report the far-field radiation patterns in the two cases. In the bare scenario, the pattern is mainly pointing towards the NSOM tip (positive z axis), with drastic reduction of scattering on the back and on the sides. This may strongly perturb the effective measurement of the nanodipole radiation features. In contrast, the pattern corresponding to the cloaked case is quasi-isotropic in the equatorial plane of the dipole with a clear dipolar shape, almost identical to that of the isolated nanodipole. It is not surprising that the bulky size of the tip may strongly affect the subwavelength nanodipole resonance, but it is impressive how a thin uniform plasmonic cloak may indeed succeed in restoring the original unperturbed radiation features of the nanodipole antenna.

As a final example of the potentials that such cloaking layers may provide for NSOM measurements, we consider the case in which the tip itself is "active," and it operates in illumination mode. Since we have proven that the cloak allows field penetration inside the cloak, at levels comparable to the uncloaked scenario, owing to reciprocity it is expected that a source placed inside the cloak may be able to efficiently radiate. In this sense, we have considered an active region at the NSOM tip, which may be obtained by coating the tip with a limited number of quantum dots or emitting molecules, as in a nano-LED for near-field illumination and sensing [4]. We consider an emitting molecule placed on the aperture of the same tip as in Fig. 1, but these concepts are also applicable to apertureless active NSOM tips operating in illumination mode (this technique would allow even smaller tips and consequently higher resolution, but the scattered fields would need to be collected by a separate sensor). In Ref. [16], we report the simulation results for the same setup as in Fig. 4, but with a passive nanodipole and an emitting molecule placed at the NSOM aperture. In the case of a bare tip (left) the strong coupling between nanodipole and tip significantly perturbs its radiation and resonance properties, and affects the dipolar shape of the near-field distribution. When the cloak covers the tip (and the optical source), excitation of the nanodipole at the same level is still preserved, but without any perturbation of its resonant properties. A clear dipolar pattern is observed right around the cloak, even though the excitation in this case lies inside the plasmonic layer.

These examples clearly demonstrate how a simple thin plasmonic layer, properly designed to cancel the scattering from NSOM tips in collection mode, may drastically enhance the imaging and scanning properties of near-field sensors in a variety of applications and operations. This may be important in a variety of scientific fields of interest, from biology, medicine, and nanotechnology to physics and optical communications.

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- [13] See supplementary material at http://link.aps.org/ supplemental/10.1103/PhysRevLett.105.263906 for additional simulation plots.Supplementary Fig. 1: Near-field plots: amplitude of the total electric field in the *H* plane (a) and of the total magnetic field in the *E* plane (b) for the bare (left column) and cloaked (right) tips of Fig. 1 for plane-wave incidence from left to right in the panels (normal incidence towards the aperture). The fields are all in scale.
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- [16] Supplementary Fig. 2 [13]: Analogous to the geometry of Fig. 4, but now feeding the nanoantenna with a nano-LED source, placed at the NSOM aperture, modeling an active NSOM tip in illumination mode, in order to excite and sense the resonant response of the nanoantenna. Amplitudes of the magnetic field on the *E* plane for the bare tip (left) and the cloaked case (right) are shown.