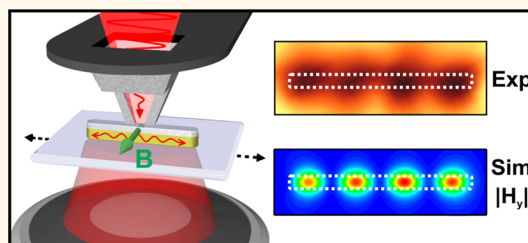


Mapping Magnetic Near-Field Distributions of Plasmonic Nanoantennas

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ABSTRACT We present direct experimental mapping of the lateral magnetic near-field distribution in plasmonic nanoantennas using aperture scanning near-field optical microscopy (SNOM). By means of full-field simulations it is demonstrated how the coupling of the hollow-pyramid aperture probe to the nanoantenna induces an effective magnetic dipole which efficiently excites surface plasmon resonances only at lateral magnetic field maxima. This excitation in turn affects the detected light intensity enabling the visualization of the lateral magnetic near-field distribution of multiple odd and even order plasmon modes with subwavelength spatial resolution.



KEYWORDS: optical magnetic field · near-field imaging · surface plasmon resonance · optical nanobar antenna · hollow-pyramid probe · probe-sample coupling

Photonic nanomaterials, and in particular plasmonic nanoantennas, enable light^{1–4} and matter⁵ manipulation at the nanoscale. They are considered to be essential building blocks for all-optical signal processing devices,^{6–10} metamaterials,^{11,12} ultrahigh sensitivity biosensors and chemical sensors,^{13–17} and active photodetectors,^{18,19} to name a few. The functionality of such photonic devices is mainly determined by the electric and/or magnetic field distributions in the vicinity of the nanostructure's boundaries. For example, in metamaterials, both the electric²⁰ and the magnetic²¹ interactions between the artificial atoms (specially designed metal nanoparticles) play a crucial role in obtaining negative permittivity and negative permeability, which are necessary for the design and the engineering of, for example, optical cloaking²² and negative refractive index materials.^{12,23} Much effort has been recently invested in achieving magnetic field enhancement at optical frequencies^{24–26} with prospective applications,²⁷ for example, as magnetic sensors^{28,29} and for achieving magnetic nonlinear effects.³⁰ Therefore, mapping both the electric and magnetic near-field

distributions has become very important for both fundamental science and applications. However, standard far-field optical microscopy methods have insufficient resolution and provide no information about the electromagnetic near-fields.^{31,32} In recent years, several techniques—each with its own specific scope, advantages, and restrictions—have been developed and optimized to gain access to the optical near-fields, for example, cathodoluminescence (CL),³³ electron energy-loss spectroscopy (EELS),³⁴ two-photon luminescence (TPL),³⁵ second harmonic generation (SHG) microscopy,⁵ and scanning near-field optical microscopy (SNOM).^{28,36–38}

In this paper we focus on scanning near-field optical microscopy (SNOM)^{39,40} where, by positioning different types of probes in the near-field of a sample, it is possible to access different components of the electromagnetic near-field. For example, the vertical (relative to the sample surface) electric field component can be mapped by the sharp needle of a scattering-SNOM.^{41,42} On the other hand, images obtained with a metal-coated optical fiber probe, having a subwavelength hole at its apex (aperture-SNOM),

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have been interpreted in terms of local density of optical states (LDOS),^{37,43,44} magnetic field,^{29,45} or lateral electric field,^{46,47} however, no consensus has been reached. While imaging of the electric field components is nowadays a standard procedure, mapping of the much more weakly interacting magnetic field components⁴⁸ remains a challenging nontrivial task. Indirect procedures, relying on the calculation of the magnetic field from the electric field *via* Maxwell's equations,^{49–52} have been suggested; however, direct measurements of the magnetic field remain difficult. Important progress in this direction has been reported by Burrese *et al.*,²⁸ who have developed a special split-ring-like probe for mapping the *vertical magnetic* field component.

Instead, here we demonstrate mapping of the *lateral magnetic* near-field distribution in plasmonic nanoantennas, in particular gold nanobars, by a hollow-pyramid probe aperture-SNOM (Figure 1a). This type of probes has been used, for instance, for investigating propagating surface plasmon polaritons (SPP).^{38,53–55} In those studies, however, the probe-sample coupling and the image contrast formation mechanism, which are crucial for understanding light confining effects in nanoantennas, have not been adequately addressed. In our paper, we propose such a mechanism and illustrate how it effectively results in mapping of the lateral magnetic field distribution of plasmonic nanoantennas.

RESULTS AND DISCUSSION

Near-field measurements are performed with a commercial SNOM system—WITec, alpha300 S.⁵⁶ The concept of the experiment is illustrated in Figure 1a and a detailed description of the setup is given in the Methods section. In short, polarized monochromatic light is focused on the apex of a SNOM probe. This probe consists of a hollow SiO₂ pyramid, mounted on an atomic force microscopy (AFM) cantilever. The pyramid is coated with a 100 nm thick Al layer, which is etched through at the apex. This opens up an aperture with a subwavelength diameter of nominally 100 nm, which provides optical resolution below the diffraction limit. For clarity, in Figure 1a, the pyramid is partially cut open. A part of the incident light can tunnel through the aperture and interact with the sample. The transmitted light is then collected and sent to the detector. Excitation and detection axes are kept collinear while the sample is being scanned in AFM contact mode underneath the probe.

The detected image contrast is a result from the coupling between the near-fields of the probe and the respective near-fields of the sample. Therefore, we first performed a separate study of the probe and the sample. Finite-difference time-domain (FDTD) simulations of the probe reveal that an incident polarized plane wave induces a dipolar charge polarization at its apex—indicated with plus and minus signs in

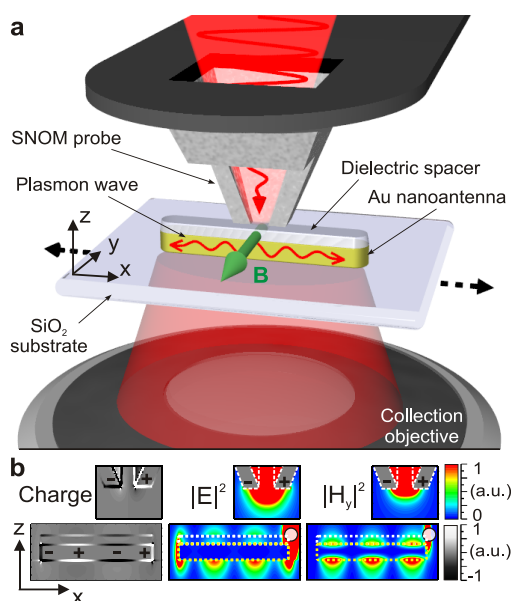


Figure 1. Near-field scanning of a plasmonic nanoantenna with a hollow-pyramid probe. (a) Schematics of the transmission SNOM setup. Electromagnetic coupling between the Al-coated hollow-pyramid SNOM probe and the gold nanoantenna induces a lateral magnetic dipole and can excite surface plasmon resonances (SPRs) in the bar. By detecting the transmitted light intensity, while scanning the sample, this dipole allows mapping of SPR magnetic field antinodes. (b) Charge density, electric and magnetic field distributions. Top: at the probe aperture for an *x*-polarized incident plane wave at $\lambda = 1 \mu\text{m}$. Bottom: for the $l = 3$ SPR antenna mode ($\lambda = 1.270 \mu\text{m}$), excited *via* an *x*-polarized dipole source, indicated with the white circle. Dimensions of the nanobar are length $L = 1120 \text{ nm}$, width $W = 70 \text{ nm}$, and height $H = 50 \text{ nm}$. The thickness of the dielectric spacer is 30 nm . *x*-*z* cross sections are taken through the probe/sample center.

Figure 1b, top. This leads to a concentration of the electric and magnetic fields, as illustrated for $|E|^2$ and $|H_y|^2$. Because of symmetry considerations, the latter is the only nonzero magnetic field component in the provided *x*-*z* cross-section, through the middle of the probe. All field components, as well as the charge and current density, near the probe aperture are given in the Supporting Information, Figure S2. The profiles are obtained at $\lambda = 1 \mu\text{m}$ and remain almost unchanged throughout the experimental spectral range. Details on the performed simulations can be found in the Methods section.

The nanoantenna structure consists of a 50 nm thick and 70 nm wide gold nanobar, covered by a 30 nm thick dielectric layer (see Methods section and Supporting Information, Figure S1). This layer enables scanning of the sample in contact mode while preventing strong conductive coupling between the probe and the sample. When such a nanobar is illuminated with light, polarized along its long axis, charge density waves at the surface of the metal are excited. They can form standing wave-like Fabry-Pérot resonances, known as surface plasmon resonances (SPRs).⁵⁷ Here, the resonance mode index l is defined

as the number of half plasmon wavelengths $\lambda_p/2$ that fit the antenna cavity at resonance. At positions with high charge density in the bar, strong enhancement of the electric near-field occurs and complementary magnetic maxima appear. This is illustrated in the simulation shown in Figure 1b (bottom) for the $l = 3$ SPR mode, where the mode index is identified from the presented near-field profiles. Here, the antenna is excited by an x -polarized electric dipole, indicated with the white circle. Again, it is worth noticing that H_x and H_z are zero in the central $x-z$ cross-section, as a result of the system's symmetry. Furthermore, the excitation of an SPR leads to resonant enhancement of the antenna's absorption and scattering cross sections.

Now that the sample and probe have been characterized individually, the near-fields induced at the apex of the probe can be used to locally excite SPRs in the near-field region of the nanoantenna. Experimental near-field scans of a single antenna with length $L = 1120$ nm measured at different excitation wavelengths are shown in Figure 2a. Normalized line scans from the maps in panel a, taken through the center of the bar, are shown in panel b. For each mode, the baseline was subtracted in order to set the transmission minimum to zero, after which the data were normalized to the transmission intensity at the substrate to account for the wavelength dependence of the probe's transmittance.⁵⁸ This normalization is chosen to optimize the image contrast, at the expense of losing the absolute transmission intensity information. The light transmitted by the probe is not fully suppressed (*i.e.*, not reaching zero) at the transmission minima. Since the nanoantenna is optically thin and narrower than the probe aperture, in addition to the finite extinction cross-section of the SPR resonances, a certain amount of light still reaches the detector. The outer boundaries of the bar are indicated with dashed lines. Well-defined dark spots corresponding to suppressed

transmission are observed, while the number of dark spots increases toward shorter wavelengths of the excitation light. From left to right, the strongest transmission contrast was obtained at $\lambda = 1480, 1000, 880,$ and 740 nm, respectively. For an accurate determination of these resonant wavelengths, near-field maps were taken in steps of $\Delta\lambda = 20$ nm and compared. The characteristic features of the experimental near-field images are similar to those of the simulated SPR near-field profiles in Figure 1b and to previous reports with scattering- and optical fiber aperture-SNOM on nanorods.^{36,37,59} The exact relation between the transmission contrast and SPR near-field distribution is, however, not straightforward, and one has to be careful with the interpretation of the SNOM images. We therefore performed extended simulations, including the probe-sample interaction, to reproduce the experimental maps and elucidate the contrast formation mechanisms.

The simulated transmission scans corresponding to Figure 2b taken at, from left to right, $\lambda = 1740, 1245, 960,$ and 810 nm, are shown in panel c. These curves were obtained by scanning the probe in 50 nm steps across the antenna while collecting the transmitted light intensity through an area spanning the same angle as the numerical aperture (NA) of the objective in the measurement. The simulated scans accurately reproduce the experimental number of transmission minima and their relative positions. The spectral shifts observed between the experimental (panels a and b) and simulated (panel c) transmission resonances most likely originate from deviations from the ideal sample and probe geometry (dimensions, shape, surface roughness, grain boundaries), as well as their optical material properties (depending on fabrication process), used in the simulations. As these parameters are hard to control and accurately determine experimentally, it is difficult to avoid such a spectral mismatch.

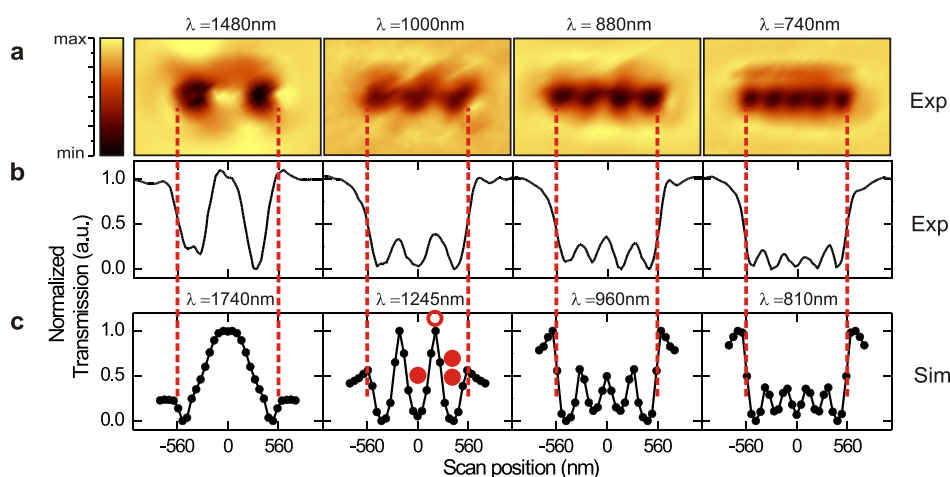


Figure 2. Experimental and simulated near-field transmission scans. (a) Experimental SNOM maps of a nanobar antenna with $L = 1120$ nm at different wavelengths. Dark regions indicate reduced transmission intensity. (b) Normalized line scans of the maps in panel a through the center of the antenna. (c) Simulations reproducing the experimental data from panel b. Red dashed lines indicate the antenna borders. Red dots and circles refer to the probe positions, discussed in Figure 3.

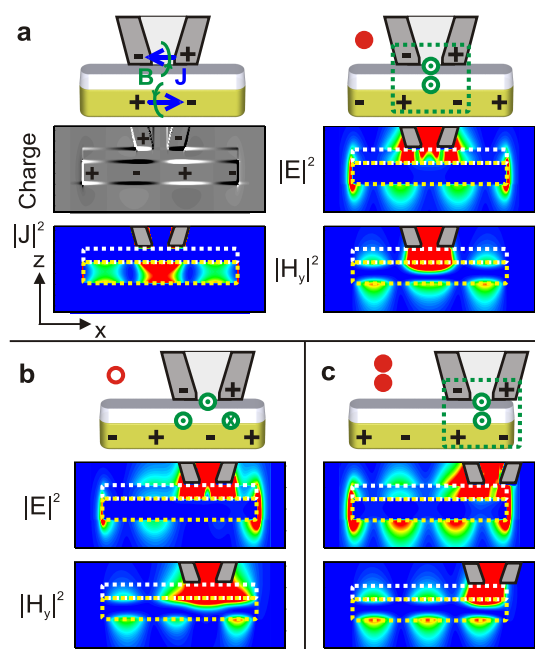


Figure 3. Probe-antenna coupling leads to efficient excitation of SPR only at the lateral magnetic field maxima. (a) Top: Illustration of image charge formation, induced current J and magnetic field B . Bottom: simulated charge, current density, $|E|^2$ and $|H_y|^2$ profiles in x - z plane through the middle of the probe and antenna at $\lambda = 1245$ nm with probe at the center of the bar. (b) Simulated $|E|^2$ and $|H_y|^2$ profiles under the same conditions as in panel a, but with probe positioned at 200 nm from the center and (c) near the edge of the antenna. Plus and minus signs indicate positive and negative charge accumulation, respectively. Green circles indicate out-of-plane magnetic field concentration. In the different panels $|E|^2$ and $|H_y|^2$ profiles have the same color scale, respectively. Red dots and circles indicate the position of the tip, shown in Figure 2c. For clarity, the schematic images are not to scale.

We can now confidently use the full-field three-dimensional simulations to investigate how the probe fields excite surface plasmons in the nanoantenna, and how this excitation influences the detected transmission intensity. Figure 3 provides a more in-depth analysis for the case where three transmission minima are observed in Figure 2c ($\lambda = 1245$ nm) and the probe is located at the same positions as those indicated with red dots and the open circle. In Figure 3a, the probe is positioned above the antenna center and the top left panel illustrates how the polarized charges at the rim of the probe generate image charges in the metallic nanostructure. This effect is evidenced by the strong electric field intensity $|E|^2$ observed between the Al coating of the probe (gray areas) and the nanobar (also seen in Figure 3b,c). This antiparallel dipole coupling effectively generates an out-of-plane y -oriented magnetic dipole (green arrows in Figure 1a and Figure 3).^{60,61} The strong lateral magnetic field enhancement is clearly observed in the corresponding $|H_y|^2$ field profiles. The charge separation induced by the probe, disturbs the free electron gas in the antenna and launches a surface plasmon wave. Even when driven at one of the SPR frequencies, the

probe couples to an SPR only at positions for which the waves, reflected back from the antenna edges, are phase matched to form a standing wave pattern. These positions are exactly the nodes in the corresponding SPR charge density distribution.

The charge distribution in Figure 3a clearly shows the standing wave pattern expected for the $l = 3$ antenna mode, exhibiting three charge nodes and four antinodes in the electric field profile. This resonant charge oscillation translates into three regions of high current density $|J|$ and consequently, through Ampère's law, three magnetic field antinodes. The top right panel of Figure 3a illustrates how the charges and magnetic near-field (green circles) at the probe aperture line up with the charge distribution and magnetic near-field of the SPR mode. When resonantly excited, both the absorption and scattering cross sections of the nanoantenna are drastically enhanced. This means that part of the power transmitted by the probe is absorbed and redirected by the antenna, resulting in a lower detected signal, and therefore, a dark spot in the SNOM image (Figure 2c, red dot).

When the probe is gradually moved off-center, the symmetric standing wave profile, clearly seen in the $|E|^2$ and $|H_y|^2$ profiles at the bottom surface of the nanobar, is disturbed. Up to a point where one of the four electric field maxima, characteristic for the $l = 3$ mode, disappears. This situation is shown in Figure 3b where the probe is shifted 200 nm from the bar's center. Here, no efficient coupling between the different electric and magnetic field components occurs, and excitation of the $l = 3$ mode is not expected. The open red circle in Figure 2c indicates that this results in a transmission maximum. When approaching the antenna edge (Figure 3c), however, the next $l = 3$ SPR magnetic antinode is probed. The symmetric SPR field profiles are restored and another transmission minimum is detected (Figure 2c, double red dot). The closer the probe is to the maximum of the antenna's magnetic field, the stronger the resulting out-of-plane coupled magnetic dipole becomes, leading to stronger excitation of the SPR mode. Consequently, more light is absorbed and less light reaches the detector. From this analysis we can conclude that the SPR field component, effectively mapped in this type of near-field measurement, is H_y .

To further illustrate that the experimental maps indeed correspond to the $|H_y|^2$ field profiles, a comparison between the measured SNOM map and the different components of the electromagnetic near-field of the nanobar is shown in Figure 4. For the simulation, the surface plasmon resonance in the antenna is excited with a dipole source, positioned above one end of the antenna. The profiles are taken at the resonant wavelength for the $l = 3$ SPR mode ($\lambda = 1270$ nm). The electric field components have a common color scale. The magnetic field components

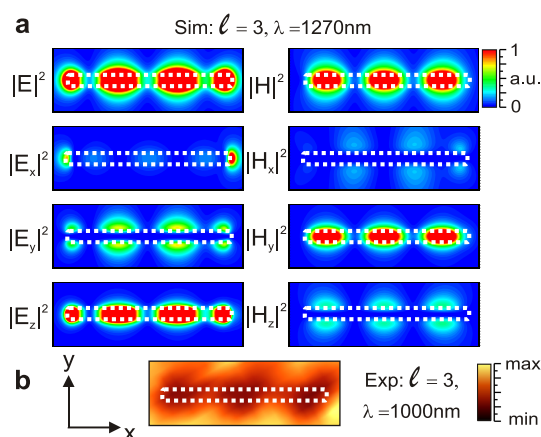


Figure 4. (a) Electric (left) and magnetic (right) field distributions for a gold nanobar with $L = 1120$ nm for the $l = 3$ SPR mode ($\lambda = 1270$ nm). x - y cross sections are taken at 30 nm from the gold top surface. (b) Experimental transmission map for the same mode reproduces the $|H_y|^2$ distribution. The white boxes outline the nanobar.

also have a common scale bar, except $|H_x|^2$ for which the maximum intensity is an order of magnitude lower.

Since a transmission minimum was shown to correspond to a region with enhanced magnetic field, it is now possible to unambiguously assign the proper mode index l to the different SPR modes seen in the SNOM maps of Figure 2. Clearly, the experimental near-field maps reproduce the calculated magnetic field maxima for the $l = 2, 3, 4,$ and 5 antenna modes, as illustrated in Figure 5. It is interesting to note that even order modes are also detected.¹³ The localized excitation of SPRs offers the necessary symmetry reduction to excite these otherwise dark modes. The simulated profiles here are also obtained by placing a dipole source above one end of the antenna to introduce the phase retardation, which however, leads to a small asymmetry seen in the profiles. From a comparison of the simulation with (Figure 2c) and without (Figure 5 bottom row) the SNOM probe, it can be concluded that the presence of the probe in the near-field of the sample leads to a slight blue-shift of a few percent of the resonant wavelengths. Spectral shifts on the same order of magnitude were observed in other SNOM

studies using different types of metal-coated and nonmetallic probes.^{62,63}

A typical near-field scan of a nanoantenna array of increasing length L is shown in Figure 6a. Following the dashed arrows, L varies from $L = 720$ nm in the top left to $L = 1800$ nm in the bottom right corner. The scan was performed in contact mode at a wavelength of 1100 nm. As L increases, additional half SPP wavelengths can fit the antenna cavity, higher order SPR modes are excited, and the number of transmission minima increases. Since the SPR modes are spectrally relatively broad, several antennas show the same number of dark transmission spots at a fixed wavelength. To define the resonant antenna length, for a specific SPR mode, again the bar showing the strongest transmission contrast for a line scan through the middle of the bar, was chosen. The resonant antennas in Figure 6a are indicated with dashed boxes and have a length of $L = 750$ nm, 1150 nm, and 1700 nm from left to right, respectively. Note that the systematically observed gradual transition between the plasmon modes excludes possible sample imperfections as the origin of the image contrast and thus, such imperfections, if present, do not affect the presented data analysis and conclusions. Performing this measurement at different wavelengths allows to obtain the SPR dispersion curve, as shown in Figure 6b (closed symbols). The SPR wavenumber $k = 2\pi/\lambda_p$, where λ_p is the SPR wavelength, is defined as $k = \pi l/L$, resulting from the geometrical condition of a standing wave in the antenna cavity.³⁷ Here, L is the antenna length and l the mode index which was demonstrated to correspond to the number of transmission dips. In defining k a possible systematic error resulting from the nontrivial phase shift of the plasmon wave upon reflection at the bar edges is ignored. This phase shift is introduced by the complex refractive index of the dispersive metallic medium of the plasmon wave.^{33,37} After measuring all points, the first few points and a few random points were remeasured to exclude possible resonance shifts caused by wearing out of the tip.

The solid and dashed lines in Figure 6b are the light lines in vacuum and in a glass medium, with refractive

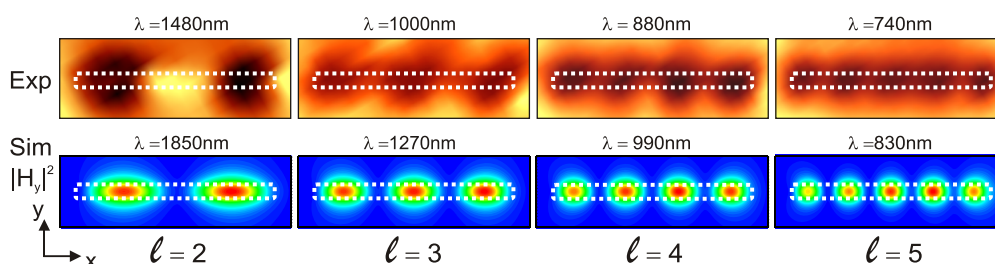


Figure 5. Experimental SNOM transmission maps reproduce the simulated SPR lateral magnetic near-field distributions. Experimental transmission minima seen in Figure 2a, zoomed-in in the top row here, correspond to the simulated magnetic near-field maxima of the $l = 2, 3, 4,$ and 5 SPR modes in the nanobar antenna ($L = 1120$ nm), shown in the bottom row. Field profiles are taken at 30 nm from the antenna surface. The white boxes outline the nanobar.

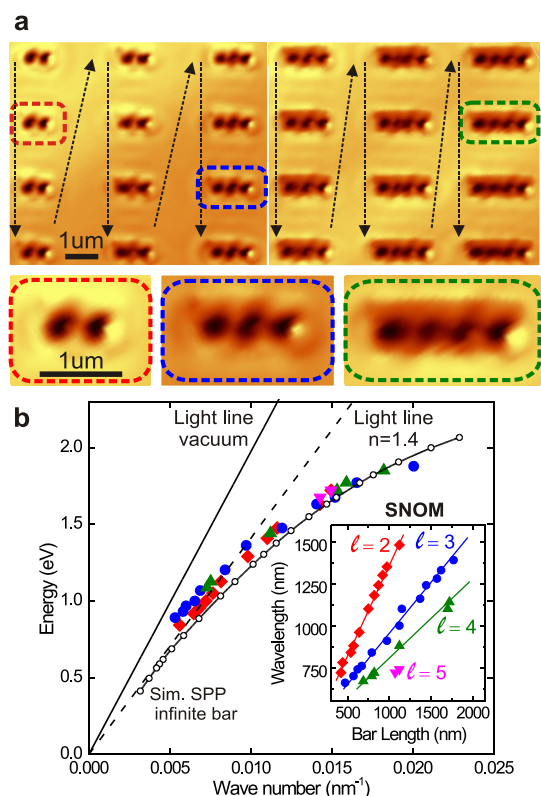


Figure 6. The dispersion relation confirms that SNOM results are related to plasmonic effects. (a) Experimental SNOM scans at $\lambda = 1100$ nm and polarization along the bars' axis of two arrays with bars of increasing length (indicated by the dashed arrows). Zoomed-in bars with resonant transmission contrast are shown in the bottom panels. $L = 750$ nm, 1150 nm, and 1700 nm from left to right, respectively. (b) Dispersion curves showing resonant plasmon energy versus plasmon wavenumber k defined as $k = \pi/L$. Closed symbols: experimental near-field transmission resonances for $l = 2, 3, 4$, and 5 modes. Solid and dashed lines show the light line in vacuum and in $n = 1.4$ medium, respectively. Black circles: calculated plasmon dispersion of an antenna of infinite length L . Inset: the dispersion data displayed as $\lambda(L)$ illustrates the expected linear wavelength scaling behavior for metallic nanorod antennas.

index $n = 1.4$, respectively. Open circles represent the calculated mode dispersion of propagating SPPs in the nanoantenna geometry with infinite length L (see Methods section). Both the experimental cavity mode dispersion and calculated SPP dispersion bend to the right of the light line at larger wave numbers illustrating the subwavelength nature of surface plasmons. This excellent agreement between the dispersion curves further demonstrates that the experimentally observed near-field transmission contrast is indeed mapping plasmonic modes in the nanobar antennas and so justifies the standing wave description.⁶⁴ The inset in Figure 6b shows the dispersion data as wavelength versus bar length. This graph further illustrates the expected linear wavelength scaling behavior for metallic nanorod antennas.^{65–67}

Finally, the presented results can be situated among other reported techniques for obtaining the magnetic

field of light. As already pointed out in the introduction, several methods to calculate the magnetic field from the electric field via Maxwell's equations have been developed in the terahertz^{51,52} and optical frequency regions.^{49,50} Although this approach has the advantage of providing information on both the electric and magnetic field components simultaneously (including their phase) it remains as an indirect method which requires postprocessing of the data. One of the most direct ways to measure the optical magnetic field is based on a split-ring probe.²⁸ This is also a phase sensitive method with, in principle, no restrictions concerning the type of optical waves under investigation. However, it has the disadvantage that such probes are not commercially available and they can only access the vertical component of the magnetic field. Devaux *et al.*⁴⁵ have reported that excitation of resonant circular plasmons in metalized aperture probes can lead to imaging of the magnetic field intensity in dielectric samples. The method is based on resonant effects in the probe and is therefore strongly restricted in terms of illumination wavelength. Another method, allowing simultaneous imaging of the vertical magnetic and electric field components in photonic crystal cavities has recently been reported.^{68,69} It is based on a particular blue-shift induced in the resonant frequencies of those samples and its applicability to other photonic (including plasmonic) systems has not been demonstrated so far.

Most notably, in contrast to the methods mentioned above, the technique we propose here provides information about the lateral magnetic field distribution in plasmonic antennas. Our measurements use commercially available probes and do not involve any additional data processing. Moreover, we expect our results to be directly applicable to the more widely used metal-coated optical fiber probes, as their geometry is very similar to the one of the hollow-pyramid probes, used in this study. Indications that the metal-coated optical fiber probes might indeed be sensitive to the magnetic near-field have been reported.^{29,45} Compared to the optical fiber, the hollow-pyramid probes are very robust⁷⁰ (we have experienced that a probe can easily provide good quality images for more than 2–3 months). The probes allow measurements in a broad wavelength region, in our case 500–1600 nm, the limitations coming from the excitation sources and the detectors. As of today, our microscope is not equipped to perform phase-sensitive measurements, but this could be implemented. It should be emphasized that the current results only demonstrate the applicability of our technique to plasmonic waves in metallic nanobar antennas. Nevertheless, imaging of the lateral magnetic field, in this work, is possible due to the antiparallel dipole coupling between the separated charges in the probe and the sample, which leads to the effective formation of a lateral magnetic

dipole. Therefore, the mapping of the lateral magnetic field distribution should be possible in any sample in which such charge separation can be induced, irrespective of the specific sample geometry. We are also optimistic to envisage that this approach could be extended to propagating surface plasmon polariton waves as well.³⁸ From this point of view, we believe that our method can be considered as a complementary one to those already reported in the literature.

CONCLUSIONS

The lateral magnetic near-field distribution of surface plasmon resonance modes in optical nanobar antennas was visualized by means of scanning near-field optical

microscopy. The formation of an effective magnetic dipole between the hollow-pyramid probe and the antenna was shown to excite standing wave-like surface plasmons in the antenna, only at the SPR lateral magnetic field maxima. This excitation results in a measurable modulation of the transmitted light intensity. These findings are of paramount importance for achieving a complete characterization, including the magnetic field components, of electromagnetic near-field light phenomena mediated by nanoplasmonic devices. Aperture-SNOM can now be considered as an important complement to the available scattering-SNOM techniques. It would, for example, be the method of choice for near-field studies of optical magnetic field enhancing and confining nanoantennas.^{24–26}

METHODS

Experimental Setup. A supercontinuum white light (SCWL) laser (NKT Photonics, Koheras SuperK Extreme Standard) with spectral range of 400–2000 nm is used as a light source. From the broad spectrum, single wavelengths with a line width of ~5 nm to ~20 nm and power of a few milliwatts, depending on the wavelength, are selected with an acousto-optic tunable filter (AOTF) (NKT Photonics, SpectraK Dual). The selected illumination is coupled into the microscope where it passes through a polarizer and is then focused on the apex of the SNOM probe by a 20 \times objective with numerical aperture NA = 0.4. The probe consists of a hollow SiO₂ pyramid, covered with an Al layer with 100 nm thickness, which is etched through at the apex. This opens up an aperture with a subwavelength diameter of nominally 100 nm, which provides optical resolution below the diffraction limit. The light transmitted through the sample is collected in the far-field by a 60 \times achromat microscope objective (NA = 0.8), directed through an analyzer, and confocally picked up by the pinhole of an optical fiber. The collected light in the fiber is delivered to two spectrometers, equipped, respectively, with a front-illuminated Si CCD camera for the visible, and an InGaAs detector array for the near-infrared spectral range. The sample is scanned in atomic force microscopy (AFM) contact mode with an XYZ piezostage, and beam-deflection laser feedback. Schematics of the experimental setup and more detailed information can be found in the Supporting Information.

Sample Fabrication. The sample consists of a 150 μ m thick glass slide coated with 10 nm of indium tin oxide (ITO) and a thin Ti adhesion layer. A 50 nm thick gold film was sputtered and covered with another Ti adhesion layer and a negative tone hydrogen silsesquioxane (HSQ) resist. The nanostructures were further structured using electron beam lithography and Xe ion milling. The resulting bar width W is ~70 nm. A residual resist layer with an estimated thickness of 30 nm remains on top of the gold particles and is not removed. Optically, this layer behaves as silicon dioxide and causes the SPR modes to redshift (e.g., 20 nm for the $l = 3$ mode of an $L = 1120$ nm antenna) due to the increase in surrounding refractive index. Additionally, the thickness of the layer will affect the spatial resolution⁷¹ and the probe-sample coupling strength.⁶³ Although a detailed study of these effects for the present system has not been performed, the results are expected to be qualitatively valid for a broad range of layer thicknesses. Most importantly, the layer serves as a dielectric insulator, preventing electrical contact, and therefore conductive coupling, between the nanoparticles and the metallic SNOM probe. This allows us to do fast contact mode scanning of the sample without drastically altering the plasmonic properties. The sample is organized in arrays, which consist of bars with increasing size in steps of 20, 30, or 50 nm.

FDTD Simulations. Simulations were performed with a commercial FDTD solver.⁷² For the near-field simulations, the nanostructure

(including the resist layer on top of the metal), the hollow pyramid, and the substrate were placed in an $7 \mu\text{m} \times 7 \mu\text{m} \times 3.5 \mu\text{m}$ volume with perfectly matched layer (PML) boundaries and a mesh of $5 \text{ nm} \times 5 \text{ nm} \times 2.5 \text{ nm}$ covering the nanostructure and the pyramid's aperture. For excitation, a plane wave source is positioned inside the pyramid. To reproduce the experimental conditions as accurately as possible, the NA of the collection objective was taken into account by calculating the transmission intensity through a rectangular surface spanning the same collection angle. The permittivity of Au and Al was taken from refs 73 and 74, respectively. The refractive index of the substrate and resist layer was set to $n = 1.4$. The ITO and Ti layers have a negligible effect on the results, and this effect is not included in the simulations. Field profiles in the absence of the probe were obtained using a point dipole source positioned above the edge of the bar in order to excite both odd and even order modes through phase retardation effects.⁵⁷ For the calculation of the plasmon dispersion in Figure 6b the mode solver of Lumerical FDTD was used.

Conflict of Interest: The authors declare no competing financial interest.

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Supporting Information Available: More details of the experimental setup and additional simulations: charge, current, electric and magnetic near-field distributions in the vicinity of the probe. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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