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Citation: Appl. Phys. Lett. 101, 131102 (2012); doi: 10.1063/1.4754534
View online: http://dx.doi.org/10.1063/1.4754534
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Real-space mapping of nanoplasmonic hotspots via optical antenna-gap loading

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(Received 30 July 2012; accepted 5 September 2012; published online 24 September 2012)

Plasmonic hotspots located in the nanogaps of infrared optical antennas are mapped in the near-field. The enhanced evanescent field resonance is shown to depend strongly on excitation wavelength, the excitation and detection laser polarization, and gap size. In addition, we demonstrate that in nanogap hotspot imaging using scattering probes, the probe tip can be considered as a load in the gap of the antenna, and the impedance of the load can then be tuned from inductive to capacitive or vice versa by changing the dielectric value of the tip load. Experimental results are in agreement with finite-difference time-domain simulations.

The experimental setup is a commercial s-SNOM system (NeaSNOM, neaspec.com), the details of which have been well described before. In short, a probing CO₂ laser is linearly S or P polarized and is focused on the tip-sample interface at an angle ~50° from the sample surface. The scattered field is acquired using an interferometric detection scheme. Suppression of background signal is achieved by vertical tip oscillation at the cantilever’s mechanical resonance frequency ω ≈ 285 kHz and demodulation of the detector signal at higher harmonics nω (n > 2). The combination of the scattered field from the tip and the reference beam pass through a linear polarizer, which selects the polarization of the measured signal either in the P or S polarization for analysis.

The Au infrared (IR) rod dipole antennas are fabricated using electron-beam lithography. They consist of approximately 17-nm-thick Au on top of a 3 nm Ti adhesion layer. Each constituent Au rod in the dimer is designed to have a width of 20 nm. Rod lengths ranging from 1.35 μm to 1.85 μm and gap sizes ranging from ~20 nm to 80 nm are fabricated on the sample. Each rod in a pair has similar geometrical parameters to within a few nanometers deviation due to inherent variations in the e-beam lithography process.

Figs. 1(a)–1(e) show near-field amplitude and phase images of an Au plasmonic dipolar antenna with gap of 30 nm, imaged at ω = 952 cm⁻¹ by implementing S/P and S/S polarization detection (we refer to the first letter as the polarization of the excitation laser and the second letter as the detection polarization throughout the text). Figs. 1(b) and 1(d) show S/P polarization-specific optical amplitude image (Fig. 1(b)) and phase image (Fig. 1(d)). The amplitude image displays a stronger optical contrast at the ends of the arms of the rods and weaker contrast at the gap region and in middle region of each rod, whereas the phase image shows the ends of the rods at 180° phase difference. When S/S polarization detection is used, the amplitude image (Fig. 1(c)) shows similar contrast as S/P on the rod ends; however, the gap region shows strong optical contrast. The phase image (Fig. 1(e)) displays similar contrast at the ends of the rod and in the gap.
region. These optical images confirm polarization-dependent dipolar plasmon excitations.\textsuperscript{11,12} Figs. 1(f)–1(j) show high-resolution S/S near-field amplitude and phase images of the gap region obtained at two different laser frequencies. The dipolar field intensity decreases both in amplitude and phase in the gap region as the laser frequency is tuned from the resonance frequency of $952 \text{ cm}^{-1}$ (Figs. 1(h) and 1(j)) to $978 \text{ cm}^{-1}$ (Figs. 1(g) and 1(i)), revealing strong wavelength-dependent excitation in accordance with FDTD simulations (supporting material\textsuperscript{16}).

Fig. 2 shows a zoom-in of the 4th harmonic near-field amplitude and phase images of hotspots at varying gap widths obtained using laser frequency, $\omega = 952 \text{ cm}^{-1}$ and implementing an S/S polarization detection scheme. The amplitude images show increasing contrast localized in the gap region as the width of the gap decreases (Fig. 2(e)), which is a manifestation of stronger localization of the dipolar field of the rods as their spatial separation diminishes. The dotted lines in Fig. 2(e) are guide to the eye. The phase images show similar contrast across the arms of the rods as reported before for S/S polarization configuration with a slightly stronger phase contrast at the gap region.\textsuperscript{13}

To understand the hotspot imaging mechanism and the effect of the probe tip on gap field formation, we performed imaging experiments using a metallic tip for comparison to imaging performed using a Si tip. In Figure 3, we show near-field amplitude and phase images of the hotspot using Si tip (Figs. 3(b) and 3(d)) and metallic tip (Figs. 3(c) and 3(e)) using S/S excitation and detection at frequency, $\omega = 952 \text{ cm}^{-1}$. The amplitude image taken using the Si tip shows the expected hotspot in the gap field. But this hotspot is absent in the amplitude image taken using a metallic tip. The phase is the same in both arms of the antenna in both images, as expected from S/S polarization imaging; however, the image taken using a Si tip (Fig. 3(d)) shows a similar phase contrast in the gap region while the image taken with metallic tip does not show phase contrast in the gap region. Since the two images were taken keeping all experimental parameters similar except for the change of tips, we conclude that the dielectric constant of the tip plays a major role in imaging antenna gaps in s-SNOM. Experiments performed on larger gap sizes to assure the metal-coated tip accesses the gap region yield the same result (supplemental information\textsuperscript{16}).
be shown to strongly depend on the dielectric constant of the tip load. Based on this, we can theoretically show this effect by using the model approximation we made above where the gap is filled with a Si or Pt load as a rod placed in the gap region to simulate the tip. The impedance of the load can then be calculated using $Z_{\text{load}} = \frac{g}{\omega \epsilon_{\text{load}} A}$, where $g = 50 \text{ nm}$ is the gap width, $A = 32 \text{ nm} \times 95 \text{ nm}$ is the cross-sectional area of the load and $\epsilon_{\text{load}}$ is the dielectric constant of the load at $952 \text{ cm}^{-1}$ ($\epsilon_{\text{Si}} = 10.2 + 8.35i$ and $\epsilon_{\text{Pt}} = -1000 + 500i$). Using the above equation, we find the load impedance for Si and Pt in the gap to be $830 + 1014i$ and $6.91 - 13.82i$, respectively.

Comparison of the load impedances with the intrinsic impedance of the dipole antenna, which is calculated to be $28 + 9.6i$, shows that the load impedance is much larger than the dipole impedance for the Si load filling the gap, whereas the Pt load is comparable to dipole impedance. The large Si load impedance means that the load increase the capacitive effect of the antenna, resulting in large charge accumulation in the gap region, which is responsible for the hotspot observed when imaged using a Si tip. On the other hand, the load resistance is smaller compared to the dipole impedance when a metallic tip is loaded in the gap region, decreasing the accumulation of charges (i.e., screening), which leads to less intense field in the gap region.

Noble metal loads are inherently inductive, and the load impedance generally depends on the load’s dielectric values (Fig. 3(k)). Fig. 3(k) shows a contour plot of the imaginary ($\text{Im}(\epsilon_{\text{load}})$ vs. real ($\text{Re}(\epsilon_{\text{load}})$) parts of the dielectric constant ($\epsilon_{\text{load}}$) of the rod material filling a 50 nm nanoantenna gap, with the color corresponding to the reactance of the antenna. The figure shows that the reactance of the load becomes inductive or capacitive based on the sign of the $\text{Re}(\epsilon_{\text{load}})$. We also note that the conclusion does not change if there is an air gap between the load and the antenna rods (supplemental material), as shown by FDTD calculations in Figs. 3(f)–3(i). Although there exist fields in the gap region between the load and the antenna, these fields are not detected. Since the s-SNOM tip scatters the enhanced field locally at the position of the load (tip), there will be no field that can be detected using an inductive load (tip). Thus, we conclude that the absence of the near-field distribution when using Pt tip is a result of the change of the dielectric value of the tip load from capacitive to inductive load impedance. Also, we note that the presence of a metallic tip shifts the resonance away from the laser wavelength towards the blue compared to Si tip (supplemental information). However, the shift is very small, and the signal in the gap still remains much weaker compared to Si tip in the entire spectral region, confirming our conclusion that the main reason why the metal-coated tip does not give correct hotspot images is because of the inductive loading effect of the tip.

In conclusion, we have employed s-SNOM to map the near-fields surrounding the nanoscale hotspots of Au IR dipolar antennas. By adjusting the polarization of the detected light, we measure both the in-plane and out-of-plane near-field components and show that the in-plane field strength increases as the gap size decreases. More importantly, we demonstrate the influence of the tip itself on the near-field behavior of the antenna. In these experiments, the
tip acts as a reporter object, similar to a molecule in a surface-enhanced Raman scattering (SERS) hotspot, which loads the feed-gap of the antenna structure. By acting as capacitive loads with minimal loss, Si dielectric tips are shown to lead to strong in-plane fields in the gap region, whereas Pt metal tips act as inductive loads, screening charges and resulting in the observation of only small in-plane fields when the tip is located near the gap. This technique offers a potential method for tuning the load impedance of antenna structures. Loading can be altered more coarsely by changing the dielectric function of the tip, and more sensitively by adjusting the position of the tip in the feed-gap, thereby modifying the tip-antenna interactions.

We thank Z.H. Kim (Korea University) and R. Hillenbrand (CIC nanoGUNE, San Sebastián) for discussions. This work was supported by the U.S. Army Research Office, Agreement Number: W911NF-12-1-0076. Work at the Molecular Foundry was supported by the Director, Office of Science, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.


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20See supplementary material at http://dx.doi.org/10.1063/1.4754534 for additional experimental and FDTD simulation results.