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Denitza Denkova

Optical Characterization of Plasmonic Nanostructures: Near-Field Imaging of the Magnetic Field of Light

Doctoral Thesis accepted by
KU Leuven, Belgium

Springer

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Life's a journey, not a destination
"Amazing", Aerosmith

Supervisor's Foreword

As a Ph.D. student, Denitza Denkova joined our Institute for Nanoscale Physics and Chemistry at Department of Physics and Astronomy, KU Leuven, to investigate near-field optical properties of plasmonic nanostructures. In the framework of her Ph.D. research, she has been hard working and efficient in pursuing challenging problems in modern optics and, more specifically, in nanoplasmonics. In her Ph.D. work Denitza has discovered a series of new phenomena in several well-defined individual and coupled plasmonic nanoantennas. To be more specific I would like to comment on her remarkable work on visualizing magnetic hot spots in plasmonic nanoantennas.

Denitza, in collaboration with Niels Verellen, Alejandro V. Silhanek, Ventsislav K. Valev, Pol Van Dorpe, and me, has developed and realized the first direct visualization of the near-field magnetic field component (Hy) of plasmonic nanostructures by scanning near-field optical microscope (SNOM). This is clearly a major achievement in nanoscale imaging of both electric (by now a well-established technique) and also magnetic fields generated by nanoplasmonic structures. Needless to say, this finding will open new horizons in nanoscale imaging in modern nanoplasmonics.

The photonics community has long disregarded the magnetic component of the electromagnetic field of light, focusing mostly on exploiting the electric field. The reason is that at optical frequencies, natural materials do not exhibit strong magnetic response, so the magnetic field is, first, not of crucial importance for those materials, and, second, very difficult to measure. The situation changes dramatically for the new classes of artificial materials, such as optical nanoantennas, plasmonic devices, and metamaterials. They are typically designed to have a very strong magnetic response; therefore, accessing their magnetic field is of crucial importance for their exploration. While in free space, the magnetic field component can be extracted from the electric one, the confinement of the light in these novel materials prevents such a straightforward relation.

Quite remarkably, by imaging the magnetic field component, Denitza has resolved a very puzzling fundamental issue, which can be formulated in the language of a quantum mechanical "Schrödinger cat" problem: How many stripes does the "plasmonic cat" have? Surprisingly, looking at the stripes of "plasmonic cat" through an "electric field magnifying glass" (SNOM in the E- imaging mode) one can see three stripes, whereas when observing the same cat through a "magnetic field magnifying glass" one can see only two stripes. This surprising difference in the observed number of stripes turns out to reflect a profound natural difference in the boundary conditions for the electric and magnetic fields confined in the restricted geometries of plasmonic nanoresonators.

These new findings, forming the basis of Denitza's Ph.D. thesis, will have an important impact on our fundamental understanding of local magnetic and electric fields in plasmonic nanostructures, and also on the development of nanophotonic devices with promising applications in chemistry (catalysis, bio- and chemical-sensing), energy harvesting, and biomedicine (cancer treatment by hypothermia, early disease diagnostics). This work also offers a remarkable new way, different from conventional techniques, to investigate experimentally various nanostructures and also for studying materials showing strong magnetic interaction with light in the areas of metamaterials, chemistry, and all-optical chips.

I am sure that Denitza Denkova will continue to contribute very creatively to research in the fields of nanoplasmonics, photonics as well as optics and condensed matter physics in general. She is clearly a very talented young scientist who pursues scientific problems at a very high level of sophistication and I wish her all the best in her scientific carrier.

Belgium
April 2015

Prof. Victor V. Moshchalkov

Abstract

The interaction of light with nanoscale objects has been one of the hot topics in the field of photonics in the past few years. More specifically, one of the promising directions is the field of plasmonics, which deals with the interaction of light with metals. Illumination of metallic nanoparticles results in the excitation of so-called surface plasmons—collective oscillations of the free electrons in the metal, driven by the incoming light. At certain frequencies, different resonant modes can be excited, which results in significant enhancement and localization of the light in the close vicinity of the nanostructure—the particle is basically acting as an antenna at optical frequencies. Such local light field enhancements (also called *hot spots*) have shown promising applications in various areas, for example for single molecule detection, bio- and chemical-sensing, all-optical chips, cancer diagnostics and treatment, *etc*.

Further development of those applications requires detailed understanding of the full picture of the light–matter interactions. Since light is an electromagnetic wave, the response of a material to illumination is determined by the interaction of the incoming electric and magnetic fields with the medium. Thus, for the plasmonic structures, depending on the specific application, it is crucial to characterize the resonance wavelengths for the different resonant modes, the charge and current distributions in the particles, as well as the electromagnetic near-field distribution of the light in the vicinity of the structure. Different methods, each with its own scope, advantages and disadvantages, already exist for imaging most of those parameters.

Still, one of these parameters, namely the *magnetic* near-field distribution of photonic structures, has received far less attention from the optics society. The reason is that at optical frequencies natural materials interact mainly with the *electric* component of the electromagnetic field of light and negligibly weakly with the *magnetic* one. Therefore, the magnetic light–matter interactions are first, not of crucial importance for those materials and second, very difficult to measure.

However, recently new classes of artificial materials, so-called metamaterials, have been developed, which enhance and exploit these typically weak magnetic light–matter interactions to offer extraordinary optical properties. For example, such

materials could refract light in a direction, opposite to the one in all conventional materials. This can allow realization of exotic devices, such as an invisibility cloak, flat lens with no resolution limit, *etc*. Therefore, in the last years, a need has appeared for both optical magnetic sources and for detectors of the magnetic field of light and tremendous efforts have been invested in this direction.

The main goal of this Ph.D. is to offer new experimental possibilities for near-field imaging of the magnetic field of light with sub-wavelength resolution and to implement the technique for studying different plasmonic nanostructures.

The experimental technique which we use is scanning near-field optical microscopy (SNOM). This is a method, based on the scanning of a probe in the near-field of the sample. Depending on the type of probe and measurement configuration, different field components can be accessed with sub-wavelength resolution. Imaging of the different *electric* field components is nowadays a well-developed procedure. However, it is still an ongoing challenge to experimentally access the *magnetic* field components, which interact much weakly with materials. Recently, it has been shown that the *normal* (relative to the sample surface) magnetic field component can be accessed by a split-ring aperture probe.

To fill in the last standing gap in the full electromagnetic mapping of the near-field of photonic devices, in our work we focus on imaging of the *lateral* (tangential) magnetic field component of the light. We demonstrate that the metal-coated SiO_2 hollow-pyramid circular aperture probe of a SNOM can be used as a detector for the lateral magnetic field of light. Moreover, we show that it can also be used as a tangential magnetic dipole source. We use the technique to study the lateral magnetic field of plasmonic structures with different geometries.

During the period in which our work has been carried out, the task for mapping the lateral magnetic field has been also addressed by other groups. They use a similar SNOM technique, which however utilizes other types of probes, based on optical fibers. The Si-based probes which we use have the advantage of being more robust and providing images with better topographical resolution, simultaneously with the optical image. A more detailed comparison is presented in the thesis.

In Chap. 1 we first discuss the interaction of light with metallic nanostructures and the resulting plasmonic effects in those structures. Several state-of-the-art applications are highlighted, which, naturally, require detailed characterization of the plasmonic structures. In the second section we explain the experimental difficulties for plasmonic studies and how the scanning near-field optical microscopy technique can overcome them. The final section discusses the physical origin behind the weakness of the magnetic light–matter interactions at optical frequencies. We also explain the specific SNOM setup which we use to probe those interactions and to image the magnetic field of light.

In Chap. 2 we experimentally demonstrate that the hollow-pyramid SNOM probe images the lateral magnetic field of light of different plasmon modes in plasmonic bars with different lengths. Supported by simulations, we describe how the coupling between the probe and the sample induces an effective magnetic dipole, which, during the scanning of the probe, efficiently excites the plasmons in the bar only at the positions of the lateral magnetic field maxima. Respectively, at

those positions the absorption in the bar is enhanced and the transmitted light is reduced. This effectively results in imaging of the lateral magnetic field of the plasmonic structure, with reduced transmitted light intensity at the positions with high lateral magnetic field. By measuring different bar lengths at different wavelengths and identifying the respective plasmon resonant modes, we construct a dispersion relation (energy vs. wavenumber) graph. The dispersion relation shows the characteristic curving deviation from the straight line of the light in dielectric media. This confirms that the effects which we observe are indeed of plasmonic nature.

In Chap. 3, supported by simulations, we discuss the theoretical background behind the imaging of the magnetic field of light with the hollow-pyramid aperture SNOM probe. Additionally to the previous chapter, here we suggest that (i) the stand-alone probe might be considered as a tangential optical magnetic point dipole source without the need to invoke the coupling to the sample, and (ii) it might be considered as a tangential optical magnetic field detector. Hence, we suggest that the probe is effectively behaving similarly to a lateral magnetic dipole. This is demonstrated for a metallic sample, while the applicability of the approximation for studying dielectric samples remains to be verified. We experimentally demonstrate the equivalence of the reciprocal configurations when the probe is used as a source (illumination mode) and as a detector (collection mode). The simplification of the probe to a simple magnetic dipole significantly facilitates the numerical simulations and the understanding of the near-field images.

Until now, we have used basic plasmonic antennas—gold bars to validate the magnetic field imaging technique. In Chap. 4, we use the validated technique to obtain the magnetic field distributions of plasmonic antennas with other geometries. We study both simple plasmonic nanoresonators, such as bars, disks, rings and more complex antennas, consisting of assembled horizontal and vertical bars in different geometrical configurations. For the studied structures, the magnetic near-field distributions of the complex resonators have been found to be a superposition of the magnetic near-fields of the individual constituting elements. This effect was confirmed by both experiments and simulations. We propose the following explanation: even though a weak interaction between the building blocks is present, the simplicity of the resonance mode structure and the clear spectral separation of the different modes in the bars result in insignificant perturbations of the near-field profile at the resonance wavelength by the presence of the additional building blocks. These findings should facilitate the study of very complex antennas, which can now be described as a simple superposition of their elementary building blocks' near-field distributions

Acknowledgments

The Ph.D. journey in not a path one walks alone, neither the Ph.D. degree is a destination one reaches alone. In the next pages, I would like to thank all the people who helped me throughout my journey and made it possible for me to reach my destination.

I would like to begin by thanking my supervisor, Prof. V.V. Moshchalkov for giving me the opportunity to come to Leuven and work on the challenging, but interesting topic of magnetic near-field imaging of plasmonic structures. Thank you for the guidance on how the modern scientific world works, but also for allowing me to preserve my personal style of work and giving me the freedom to fit in this complicated world in my own way. Thank you for giving me the opportunity to play with this big toy—the SNOM. It was already in the beginning we discovered that the SNOM is a she-microscope—one with a difficult character and a lot of mood-swings. But in the end, after 4 years, I believe I finally managed to get some understanding with her.

Next, I want to thank my co-supervisors—Dr. N. Verellen and Dr. V.K. Valev. Niels, I believe me being your first Ph.D. student was an interesting and useful experience for both of us! I am very grateful for all your help with the sample fabrication, simulations, data interpretation, and all other scientific (and not only) activities. Thanks for having the patience to guide me and help me. Ventsi, I want to thank you for your help, especially in the difficult starting period of my Ph.D. Thank you for diversifying my work and broadening it beyond the SNOM by involving me in your second harmonic generation studies.

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For good or bad, to me life means much more than work. So, now it comes the time to also thank all those people who extended my life in Leuven beyond work.

A big thanks to all the colleagues in the group who were trying to create a nice working atmosphere, sometimes even a smile on the corridor from you guys was enough to boost up my mood—Matias, Joffre, Lise, Joris, Misha, Paulo, Jun Li, Xianmei, Vladimir, Junyi, Dorin, Gufei, Johan! Thanks to my office-mates Paulo, Matias, and Jun Li—mainly for having the patience to re-direct everyone looking for me at the office to my lab-cave on the third floor. It is not because I don't like you guys that I came once per year to the office—just the SNOM was getting jealous every time I was not with her! Jian, keep it in mind, you have to be very careful with her!

I would keep a lot of warm memories also from the other colleagues from the department. Definitely from the "veterans", who taught me the rules of the game around Leuven—here I have no choice but to start with Mariela's name, and then continue with the other party-animals—Bas, Werner, Niels, Bart Raes, Cristian, Pieterjan, Sasha, Yogesh, Tom, Tomas, Jo (sorry Jo, you are already in the "veterans" category), Ataklti (you too, you have a baby already!!!), Joffre (you have always been here by default, I think). And also many thanks to "the younger guys" with whom we shared some great moments: Tobias and Claudia (you know that I am in love with the gezelligheid you create around yourselves, right?), Kelly and

Tom (I still want to try the water skiing!), Barbara and Didier, Hanna and Miro, Matias, Mattias, Piero and Pía, Bart, Johanna, Hiwa, Manisha, Sérgio, Petar, Misha, Vera, Bas, Fille, Fille, Fille, Katrien, Bart Ydens, Zhe, Winny, Cédric, and others.

Since sports is a huge part of my life, I want to also thank all the people who had the patience to teach me new sports or to get trained by me or simply the ones who dared to do sports with me.

Historically, one of my first social activities in the group was to join the soccer team—those guys from the Physics Department had a special style of making friends with ladies…. No, seriously, thanks to Jo, Ataklti, Bas, and Joffre for daring to bring women to the soccer team! It was very relaxing to kick some balls with all of you guys—Petar, Daniel, Fabian, Piero, Matias, Bert, Tomas, Pieter, Troy, Mansour, and of course the engineers—Son (thanks for organizing!!), Ruben, Dries, Tijs, Damien, Carlos, Mats, Asim, Gonzalo, Paola, Vittorio, Giovanni, Tassos, Guilherme, Sam, and all other Messi's on the field.

The other sport which you guys definitely got me hooked into was the squash of course! I know you mainly invited me to join because you were too lazy to go by bikes and you needed a transport by my limo-mafia car, but still, we reached a good synergy! Thanks for having the patience to teach me to play, I am now really a big fan! I believe the main credits here have to go to Mariela (my first opponent ever and also the first one to ever let me win), Bas (most definitely my best squash performance ever was that long-rallies-show which unfortunately ended up 3:2 for you), the Fille's (pity that you guys got scared and stopped coming just when I was ready to beat you!), Jo (I would dare to challenge you now!), Ewald, Bart, Werner, Kelly, Tobias, Arnaud; to the new generation—Carlos (! Si senõr!, I will start getting those drop shots one day), Joffre (for bringing up the competitive mood on the field), Enric (I am sure I won two sets that day!!), Matias (sorry, but I have to say it—my first, and probably only one ever 11:0 win), and the brand-new generation—Mattias, Pierro, and Bart—I am relying on you guys to keep up the squash group playing! It was also a pleasure to play with the engineers—Vyacheslav (thanks for organizing!!), Laurens, Damien, Axel, Yas, Sofie, Lucie, Bertram, and the rest.

Now of course we come to the biggest passion—volleyball! Guys, I can really not believe how hooked and enthusiastic you became for the volleyball and especially for the beach volleyball! I am really, really proud of you—you learned the triangulation, the kitty, and so many professional volleyball tricks! Thanks to all of you who shared the moments on the "beach"—Matias and Hiwa (thanks to both of you for keeping the organization running), Joffre (being as lively as you are, you are doing great regarding most of the important elements—counting the points, reserving the fields, fetching the balls and the only thing you might still work on a bit is your reception), Carlos (you are sometimes too demanding!), Bas (you are loosing shape), Mattias (that pass is not that important), Johanna, Vera, Manisha, Sérgio, Jochen, Ivan, Nastya, Stas, Ivana, Jono, and others.

And Mariela, I can really not believe it that you convinced me to participate in the Bike and Run (I knew I should have gone home after the first Chouffke) and I can definitely not believe that I liked it! Of course this was partially because of the huge euphoria in the whole department and the fantastic T-shirts we had. But mostly, it is because of my personal coach and running-buddy—Mattias. Kiddo, I really had a great time, I cannot believe that we made it in 2:43:14, 12.5 km/h!!! And hey, Ironman… come on, really… a pot on the bike?! We had the best style of all the teams for sure! Maybe only Hanna's team was competing with that picnic basket.

There are also plenty of other memorable events, for which I want to thank all participants: the crazy summer trips to the Rotselaar lake; the skiing trip to Austria, the Christmas Markt in Cologne, the motorbike trips, that legendary Hawaiian party, the cozy board game evenings at Tobias and Claudia's fantastic apartment, the countless just-one-beer visits to the Oude Markt, the once-in-a-lifetime pantoffels night, the refreshing ice cream and coffee-breaks just (but not restricted to) when it was needed. And of course the blasting Ardennen weekend (I accept betting on how many times I use the word "Thanks" in the Acknowledgments)…!!!

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Thank you all!

Best wishes,
Denitza

Contents

Abbreviations

AFM	Atomic force microscopy
AOTF	Acousto-optic tunable filter
CNT	Carbon nanotubes
EBL	Electron beam lithography
FDTD	Finite-difference time-domain
FTIR	Fourier-transform infrared spectrometer
LDOS	Local density of optical states
PALM	Photo-activated localization microscopy
PMMA	Poly (methyl methacrylate)
SCWL	Supercontinuum white light
SEM	Scanning electron microscopy
SERS	Surface-enhanced Raman scattering
SHG	Second harmonic generation
SNOM	Scanning near-field optical microscope
SPR	Surface plasmon resonance
STED	Stimulated emission depletion
STORM	Stochastic reconstruction microscopy
TEM	Transmission electron microscopy
TIRF	Total internal reflection microscopy
TOA	Tip on aperture
TPL	Two-photon luminescence

Symbols

a	Aperture diameter
A	Lens size
α	Fine-structure constant
$B = \mu_0 H$	Magnetic induction
c	Speed of light
δ_x	Spatial resolution
$\boldsymbol{\delta}_\theta$	Angular resolution
D	Distance between lens and sample
e	Elementary charge
E	Electric field
$\mathcal{E}_O$	Permittivity of free space
F_b	Magnetic force
F_e	Electric force
G	Gap size between two parallel bars
h	Bar height
H	Magnetic field
$\hbar$	Reduced Planck constant
J	Current density
k	Wave number
l	Plasmon mode number
L	Bar length
λ	Wavelength
m	Mass of a moving charged particle
μ_0	Permeability of free space
n	Refractive index
r	Distance between an aperture and a point of observation
r_A	Radius of the first Airy disk
r_B	Radius of an electron's orbit
R	Distance between an aperture and a screen

θ	Acceptance angle of an objective
q	Electric charge
v	Speed of a moving charged particle
W	Bar width