Surface Plasmon Effects on the Far-Field Signals of AgO_x-Type Super Resolution Near-Field Structure

Wei-Chih LIU*, Ming-Yaw NG and Din Ping TSAI¹

Department of Physics, National Taiwan Normal University, Taipei, Taiwan 116, Republic of China

¹Department of Physics and Center of Nanostorage Research, National Taiwan University, Taipei, Taiwan 10617, Republic of China

(Received December 5, 2003; revised January 14, 2004; accepted January 23, 2004; published July 29, 2004)

A super-resolution near-field structure (Super-RENS) of AgO_x type could perform the task of high-density near-field optical recording in a more feasible way. To further explore the optical resolution and controllability of super-RENS disks, the near-field and far-field optical properties of the AgO_x-type super-RENS embedded with random silver nanoparticles and nano cavities of different densities were studied using finite-difference time-domain simulations. The random nanostructures yielded super-resolution capability, but the surface plasmon effects generated by the random metallic nanoparticles significantly enhanced the far-field signals. A simplified Fourier optical theory was proposed to understand the relationship between the enhanced near field of random nanostructures and the super-resolution capability in the far field.

[DOI: 10.1143/JJAP.43.4713]

KEYWORDS: super-resolution near-field structure, localized surface plasmon, metallic nanoparticles, finite-difference time-domain method

1. Introduction

The super-resolution near-field structure (super-RENS) has important applications for high-density optical storage and nano photonic devices.^{1–3} Compared to the Sb-type super RENS, both the AgO_x-type super-RENS and the PtO_x-type super-RENS have stronger local field enhancement and higher carrier-to-noise ratio (CNR).^{4–7} In comparison to high-density optical storage systems achieved by conventional scanning near-field microscopy technologies,^{8,9} the super-RENS avoids the difficulties of feedback control of the near-field distance and the degrading of the near-field probe, and opens a new approach for near-field optical research.

Using the three-dimensional finite-difference time-domain (FDTD) method, Nakano *et al.*¹⁰ and Kataja *et al.*¹¹ have investigated the far-field optical properties of AgO_x-type super-RENS disks with one dissociated silver particle in the AgO_x layer. Shi *et al.*^{12,13} have also investigated a new type of the super-RENS disk especially with regard to its heating properties using numerical simulations and experiments. Previously the AgO_x-type super-RENS has been studied using near-field scanning measurements and FDTD simulations by our group.^{14–17} Multiple photodissociated silver nanoparticles were assumed to be embedded in the AgO_x layer and the collective effects of localized surface plasmons of silver nanoparticles were suggested to be the working mechanism of the super-RENS for near-field optical storage. The silver nanoparticles and their surrounding localized evanescent waves act as virtual near-field optical probes and play a role similar to the fiber tip in an apertureless near-field optical microscope. The major difference is that in the AgO_x-type super-RENS, many silver nanoparticles are embedded in the AgO_x layer and their collective surface plasmon effects can greatly enhance the reading efficiency.

Nevertheless, the far-field effects of the localized surface plasmons are still not clear. In the case of the AgO_x-type super-RENS disks, the far-field signals of the optical readout result from complicated near-field interactions between the random silver nanoparticles and the recording layer with

subwavelength recording marks. Because conventional optical simulations do not directly analyze the effects of near-field interactions, in this paper the FDTD method was used to systematically simulate the near-field and far-field optical signals from AgO_x-type super-RENS disks. Furthermore, we also compared the far-field signals of embedded silver nanoparticles with those of embedded nano cavities in the AgO_x layer to examine the surface plasmon effects on the far-field signals of AgO_x-type super-RENS disks.

2. Simulation Model

The two-dimensional FDTD method was used to systematically simulate the optical near-field distributions of the AgO_x-type super-RENS disks (see Fig. 1) with random silver nanoparticles embedded in the AgO_x layer and with recording marks of different lengths. The wavelength of the Gaussian beam was 650 nm and the numerical aperture (NA) of the lens was 0.85. The structure of the AgO_x-type super-RENS was digital versatile disk (DVD) substrate/ZnS-SiO₂ (20 nm)/AgO_x (15 nm)/ZnS-SiO₂ (20 nm)/Ge₂Sb₂Te₅

Fig. 1. Scheme of the AgO_x-type super-RENS disk with embedded random silver nanoparticles.

*E-mail address: wcliu@phy.ntnu.edu.tw

Table I. Indices of refraction at 650 nm.

Material	Index of refraction
substrate	1.5
ZnS-SiO ₂	2.07
AgO _x	2.7
Ag	0.055 + 4.44i
GST(amorphous)	3.38 + 4.07i
GST(crystalline)	3.855 + 4.80i

(15 nm), and their indices of refraction are listed in Table I. The ZnS-SiO₂ layers were protective dielectric layers and one of their functions was to retain dissociated oxygen. Recording marks were created by a high-intensity laser beam focused on the phase-change recording medium Ge₂Sb₂Te₅ (GST), and the refractive index of amorphous GST was different from that of crystalline GST. In the recording layer, the recording marks were posed periodically and the distance between recording marks was equal to the length of each recording mark. In this work, the imaginary part of the refractive index of amorphous GST was larger than usual, but it did not cause significant impact because we normalized the far-field signals.

The distribution and the density of embedded silver nanoparticles play an important role in achieving the super-resolution. To simplify the situation, the size variation of silver nanoparticles was ignored in this paper, and we concentrated on the distribution of silver nanoparticles. The diameter of each silver nanoparticle was 3 nm and the dispersion of silver was simulated using the Lorentz model.^{18,19)} The random silver nanoparticles were generated either in the fabrication process in uniform random distribution or by the photodissociation or heatdissociation process in Gaussian random distribution by an incident laser beam. Therefore silver nanoparticles were set in a combination of a uniformly random distribution and a Gaussian random distribution. Four distributions of silver nanoparticles with different densities were studied in this work. Table II shows the details of each distribution, which include the number of nanoparticles in the uniformly random distribution and that in the Gaussian random distribution. Distribution A had a total of 560 silver nanoparticles distributed in a 4800 nm computational region and had the highest density. Distributions B, C and D had 340, 120 and 50 silver nanoparticles distributed in a 4800 nm computational region, respectively. Silver nanoparticles of high density generated enhanced localized fields and excited surface plasmons, and it was reasonable to expect efficient evanescent wave coupling in high-density cases.

Surface plasmon effects only exist around metal-dielectric

interfaces. Without any metallic composition, dielectric nanostructures still exhibit local fields higher than the incident field for lightning-rod effect. The local fields surface plasmon excitation can be several orders of magnitude higher than the local fields from the lightning-rod effect. To demonstrate the surface plasmon effects, the silver nanoparticles in the AgO_x-type super-RENS disks were replaced with nano cavities of the same size and distribution. The nano cavities could induce the lightning-rod effect, but not surface plasmons.

To obtain detectable far-field signals, the reflected far-field intensity signals were calculated using the near-to-far-field transformation¹⁹⁾ from the near-field distributions. The far-field intensity differences between the on-mark and off-mark cases of the super-RENS structures were computed for various mark lengths. The on-mark case meant that the incident light focused on the recording mark, and the off-mark case meant that the incident light focused between two recording marks. The far-field intensity difference signals demonstrated the ability of the super-RENS to distinguish the subwavelength recording marks, and they were related to the resolution of the super-RENS.

3. Results and Discussion

Figure 2 shows the near-field distributions of random silver nanoparticles and nano cavities with TM-mode incident light. In distribution A with high density, silver nanoparticles formed strongly scattering centers, and localized surface plasmons were excited. In contrast, the local fields of the nano cavities were significantly weaker than those of the silver nanoparticles. Although the size and the distribution in both cases were the same, distinct differences in the local fields were due to localized surface plasmon excitation in the silver nanoparticle clusters.

For different densities of silver nanoparticles, the local field enhancements generally became lower with decreasing nanoparticle density.¹⁷⁾ The far-field signals of silver nanoparticles with different densities are shown in Fig. 3. The far-field intensity difference signal in Fig. 3 is the difference between reflected far-field signals with the laser beam focused at the recording mark and off the recording marks. Each curve was normalized to the far-field intensity without recording marks. Without silver nanoparticles, the far-field signals were indistinguishable when the mark size was less than 200 nm, i.e., below the diffraction limit. When the silver nanoparticles were presented, the far-field signals were several magnitudes higher and recording marks as small as 50 nm were distinguishable. These results clarified the physical mechanism by which the AgO_x-type super-RENS achieved resolution beyond the diffraction limit. For different densities, distribution A with high density had the

Table II. Random distributions of silver nanoparticles in a 4800 nm region.

Distribution	number of nanoparticles in Gaussian random dist.	number of nanoparticles in uniformly random dist.	total number of nanoparticles
A	500	60	560
B	300	40	340
C	100	20	120
D	50	0	50

Fig. 2. TM-mode near-field distributions of (a) silver nanoparticles and (b) nano cavities. Each nanoparticle and nano cavity had the same size and both were in random distribution A. The recording mark length was 100 nm. Localized fields were formed around the nanoparticles and nano cavities, but the local field enhancements around the silver nanoparticles were much higher than those around the nano cavities.

Fig. 3. Far-field difference intensity signals of the AgO_x -type super-RENS disks with silver nanoparticles in random distributions A, B, C, and D. The solid line indicates the far-field difference intensity signal without silver nanoparticles. High-density distribution A exhibited the strongest far-field signals and low-density distribution D exhibited the weakest far-field signals.

strongest far-field signals, and distribution D with the lowest density had low far-field signals. The far-field signals of distributions B and C were in between both extremes. The far-field signals were closely related to the densities and the

Fig. 4. Far-field difference intensity signals of the AgO_x -type super-RENS disks with silver nanoparticles and nano cavities in random distributions A and C. The solid line indicates the far-field difference intensity signal without silver nanoparticles. Generally the far-field signals of nano cavities were about one order of magnitude weaker than those of silver nanoparticles with the same distribution.

distributions of silver nanoparticles, but they were not simply proportional to the densities of the nanoparticles. For example, at mark sizes of 150 nm and 80 nm, the far-field signals of distributions A, B, and C were similar and the far-field signals of distribution D were one or two orders of magnitude weaker. Our simulations indicated direct connections between near-field distributions of silver nanoparticles and their far-field signals.

For the case of nano cavities, the weaker near field suggested the far-field signals of nano cavities were lower than those of silver nanoparticles. The simulated results in Fig. 4 agreed with this deduction. Figure 4 shows the far-field signals of silver nanoparticles and nano cavities of distributions A and C. The random nano cavity cases still yielded resolution beyond the diffraction limit, although generally the far-field signals were about one order of magnitude weaker than those of silver nanoparticles with the same distribution. The performance beyond the diffraction limit was caused by the random distribution of the nano cavities, but the weaker far-field signals resulted from the lack of surface plasmon excitation. These results could help us to understand the complicated optical phenomena of the AgO_x -type super-RENS and to improve the performance of various types of super-RENS disks. For example, a super-RENS disk consisting of a metal-free organic layer instead of the AgO_x layer was reported,²⁰⁾ and our research provided theoretical grounds for the physical mechanism of the new super-RENS containing dielectric material.

A simplified explanation based on Fourier optics of subwavelength structures^{21–23)} has been developed to explain these results.^{24,25)} The spatial spectrum of the far-field signal was the convolution of spatial spectra of the reflected light from recording marks and the optical function of the AgO_x layer. If the spatial spectrum of the recording marks was $g(k)$, the spatial spectrum of the random silver nanoparticles was $s(k)$, and the spatial spectrum of transmitted light was $t(k)$, then $t(k)$ would be proportional to the convolution of $g(k)$, and $s(k)$, i.e.,

$$t(k) \propto g(k) \otimes s(k) \quad (1)$$

It is clear that even $s(k)$ was random but had a very broad spectrum, and it still could convolute the evanescent components of $g(k)$ into propagating components in $t(k)$. In this paper, the recording layer was a periodic structure with a subwavelength period and $g(k)$ was the spatial spectrum of a subwavelength grating. Except $g(0)$, non-zero parts of $g(k)$ were evanescent components and could not propagate to the detector. Only with the $s(k)$ of a broad spatial spectrum, the evanescent component of $g(k)$ was able to be convoluted into propagating waves and was detectable in the far field.

Figure 5 shows reflected spatial spectra sampling at the boundary of the AgO_x -type layer as functions of characteristic length $l = 2\pi/k$ and represented $s(k)$, i.e., the spatial spectra of the optical functions of the AgO_x layers with different distributions. Without any embedded silver nanoparticles, the spatial spectrum dropped quickly at subwavelength characteristic lengths, and it followed that without silver nanoparticle, the far-field signal was subjected to the diffraction limit. For various distributions of silver nanoparticles and nano cavities, the spatial spectra were random and very broad. They were still significant even with a characteristic length as small as 50 nm, which was much

smaller than the wavelength of incident light. The highest density case (distribution A) in Fig. 5(a) generally had the stronger spatial spectrum, but in Figs. 5(a) and 5(b) the spatial spectrum of silver nanoparticles was about one or two orders of magnitude stronger than that of the nano cavities. The difference was related to its high local fields and collective surface plasmon effects and was consistent with the behavior of the far-field signals in Fig. 4. Although the simplified explanation neglected the vector wave properties and the multiple reflection between the layers of the super-RENS, our results elucidated that the super-RENS disks with embedded silver nanoparticles or nano cavities could distinguish subwavelength recording marks and could transfer the evanescent waves from subwavelength marks to detectable far-field signals.

4. Conclusions

In this research, the near-field and far-field optical properties of the super-RENS with embedded random silver nanoparticles and nano cavities were studied using FDTD simulations. The far-field intensity difference signals were distinguishable with recording mark sizes far below the diffraction limit, and the resolution of the super-RENS could be controlled by varying the densities of silver nanoparticles. The random nano cavities also exhibited super-resolution capability, but the near fields and far-field signals from nano cavities were apparently weaker than those from silver nanoparticles. A simplified Fourier optical theory provided a theoretical background to explain the super-resolution capability of the super-RENS disks. Our results demonstrated that the super-resolution capability mainly resulted primarily from the spatial spectrum of nanostructures; nevertheless, the surface plasmon excitation between metallic nanoparticles could significantly enhance near field as well as far-field signals. This super-RENS is relatively easy to fabricate and able to solve the difficulty of nanoscale distance control in traditional scanning near-field optical techniques. Further research will help us to design a super-RENS with superior resolution, higher CNR, and better controllability, and will also make the super-RENS a new approach to near-field optical research.

Acknowledgment

This work was supported by the National Science Council and the Ministry of Economical Affairs (92-EC-17-A-08-S1-0006) of Taiwan, Republic of China.

Fig. 5. Reflected spatial spectra at the boundary of the AgO_x layer with random silver nanoparticles and nano cavities. The random distribution of silver nanoparticles or nano cavities was (a) in distribution A and (b) in distribution C. The dashed line indicates the reflected spatial spectrum without silver nanoparticles. The characteristic length $l = 2\pi/k$ corresponds to the transverse component of the propagation vector.

- 1) J. Tominaga, T. Nakano and N. Atoda: Appl. Phys. Lett. **73** (1998) 2078.
- 2) J. Tominaga, H. Fuji, A. Sato, T. Nakano, T. Fukaya and N. Atoda: Jpn. J. Appl. Phys. **37** (1998) L1323.
- 3) T. Nakano, A. Sato, H. Fuji, J. Tominaga and N. Atoda: Appl. Phys. Lett. **75** (1999) 151.
- 4) H. Fuji, J. Tominaga, L. Men, T. Nakano, H. Katayama and N. Atoda: Jpn. J. Appl. Phys. **39** (2000) 980.
- 5) J. Tominaga *et al.*: Jpn. J. Appl. Phys. **40** (2001) 1831.
- 6) T. Kikukawa, T. Nakano, T. Shima and J. Tominaga: Appl. Phys. Lett. **81** (2002) 4697.
- 7) H. Fuji, T. Kikukawa and J. Tominaga: Jpn. J. Appl. Phys. **42** (2003) L589.
- 8) E. Betzig and J. Trautman: Science **257** (1992) 189.
- 9) E. Betzig, J. Trautman and J. R. Wolfe: Appl. Phys. Lett. **61** (1992)

- 142.
- 10) T. Nakano, Y. Yamakawa, J. Tominaga and N. Atoda: Jpn. J. Appl. Phys. **40** (2001) 1531.
- 11) K. Kataja, J. Olkkonen, J. Aikio and D. Howe: Tech. Dig. Int. Symp. Optical Memory, Nara, Japan, November 2003, We-F-07.
- 12) L. P. Shi, T. C. Chong, H. B. Yao, P. K. Tan and X. S. Miao: J. Appl. Phys. **91** (2001) 10209.
- 13) L. P. Shi, T. C. Chong, H. B. Yao, P. K. Tan, J. M. Li, X. S. Miao and X. Hu: Tech. Dig. Int. Symp. Optical Memory, Nara, Japan, November 2003, We-F-05.
- 14) D. P. Tsai and W. C. Lin: Appl. Phys. Lett. **77** (2000) 1413.
- 15) W.-C. Liu, C.-Y. Wen, K.-H. Chen, W. C. Lin and D. P. Tsai: Appl. Phys. Lett. **78** (2001) 685.
- 16) W.-C. Liu and D. P. Tsai: Phys. Rev. B **65** (2002) 155423.
- 17) W.-C. Liu and D. P. Tsai: Jpn. J. Appl. Phys. **42** (2003) 1031.
- 18) J. B. Judkins and R. W. Ziolkowski: J. Opt. Soc. Am. A **12** (1995) 1974.
- 19) A. Taflov: *Computational Electrodynamics* (Artech House, Boston-London, 1995).
- 20) T. Shima, M. Kuwahara, T. Fukaya, T. Nakano and J. Tominaga: Tech. Dig. Int. Symp. Optical Memory, Nara, Japan, November 2003, Fr-J-05.
- 21) C. J. R. Sheppard, H. Fatemi and M. Gu: Scanning **17** (1995) 28.
- 22) J. M. Vigoureux and D. Courjon: Appl. Opt. **31** (1992) 3170.
- 23) J. M. Vigoureux, F. Depasse and C. Girard: Appl. Opt. **31** (1992) 3036.
- 24) W.-C. Liu and D. P. Tsai: to be published in Scanning.
- 25) W.-C. Liu, M.-Y. Ng and D. P. Tsai: to be published in Scanning.