Deformation and Plasmon Effects of Deformed AgO_x -Type Super-Resolution Near-Field Structure

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(Received April 16, 2006; accepted May 23, 2006; published online September 7, 2006)

The deformation and plasmon effects of silver nanoparticles in a deformed AgO_x -type super-resolution near-field structure are studied using the finite-difference time-domain method. The deformed AgO_x -type super-resolution near-field structure (super-RENS) provides an additional length (or path) that is longer than the length normal to the motion of the silver nanoparticles that are located at the far edge of the bubble from the optical axis of the incident beam. The purpose of this study is to investigate the optical properties between the active layer and the incident light by varying polarization direction, wavelength, and the particle sizes of the silver nanoparticles in the near field zone. [DOI: 10.1143/JJAP.45.7228]

KEYWORDS: deformation and plasmon effects, super-RENS, finite-difference time-domain method, nanoparticles

In the quest for a higher data storage capacity, various solutions for the future generations of optical discs are under investigation. Today, a novel emergent technique, near-field optical data storage, allows the writing and/or reading of information bits which are smaller than the resolution limit of the optical head without increasing the complexity of the detecting system. Such a technique is very attractive because of its compatibility with existing players/recorders. Near-field optical data storage techniques depend on the evanescent coupling of light to the data layer of the optical storage medium to read and write data marks that are smaller than the diffraction limit of conventional optical data storage systems. In recent years, there has been increased attention given to techniques that may overcome this limit. One promising technique uses a storage medium with a super-resolution near-field structure (Super-RENS^{1,2}). The Super-RENS was introduced by Tominaga and coworkers^{3–7} to perform the task of near-field recording in a more feasible way.

An AgO_x Super-RENS structure is composed of an active layer (AgO_x) that is sandwiched between two dielectric films (ZnS-SiO_2), and this three-layer stack is deposited directly on the phase-change data layer ($\text{Ge}_2\text{Sb}_2\text{Te}_5$) of a disk. One of the dielectric layers is directly adjacent to a polycarbonate layer acting as an insulating layer; the other dielectric layer is the cover layer. If a storage medium incorporates a Super-RENS structure, the light spot that probes the data layer is smaller than the size of the focused spot that is incident on the cover layer of the Super-RENS structure. The optical aperture serves as a probe on the disk like that for scanning near-field optical microscopy (SNOM). After recording on a phase-change layer, small marks are reproduced through the aperture. The particles, instead of the aperture, function as the scattering center, which is the key to inducing the super-resolution phenomenon. The 15 nm AgO_x thin films are considered to be nonlinear optical layers which control the near-field optical aperture. Recently, several studies have explored this nonlinear near-field optical property;^{8–13} however, the influence of the deformed Super-RENS structure does not seem to be appropriately considered. The active layer (AgO_x film) of the AgO_x -type Super-RENS structure is deformed after a writing process, as observed in experimental work.^{14,15} The silver (Ag) nanoparticles spread

out in the bubble pit (deformed AgO_x film) that is, in addition, formed at the focus spot coincidentally.

To examine this nonlinear near-field optical property and match it to a realistic situation, we study the relationship between the active layer (bubble pit) and incident light for various diameters of Ag nanoparticles in the deformed AgO_x -type Super-RENS structure using two-dimensional finite-difference time-domain (FDTD) simulations. Our results indicate a close connection between the surface plasmon effects of Ag nanoparticles and the nonlinear near-field optical property observed in experiments.

Figure 1 shows bubble pit marks formed in the active layer (deformed AgO_x film) and Ag nanoparticles distributed randomly inside the bubble. We assume that the Ag particles are of the same size and randomly distributed in the deformed AgO_x layer. The distribution density of Ag nanoparticles near the top surface of the bubble pit is lower than that near the base of the bubble pit. The layer structure is, from bottom to top, polycarbonate ($n = 1.5$)/ ZnS-SiO_2 ($n = 2.25 + i0.01$)/ AgO_x ($n = 2.270 + i0.08$)/ ZnS-SiO_2 /

Fig. 1. Scheme of bubble pit marks formed at active layer (deformed AgO_x film) and Ag nanoparticles distributed randomly inside bubble. The Ag particles are of the same size and randomly distributed in the deformed AgO_x layer. The distribution density of Ag nanoparticles near the top surface of bubble pit is lower than that near the base of bubble pit. The layer structure is, from bottom to top, polycarbonate ($n = 1.5$)/ ZnS-SiO_2 ($n = 2.25 + i0.01$)/ AgO_x ($n = 2.270 + i0.08$)/ ZnS-SiO_2 /Ge₂Sb₂Te₅ (GST, crystal phase, $n_c = 4.01 + i3.61$)/ ZnS-SiO_2 /air ($n = 1$).

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$\text{Ge}_2\text{Sb}_2\text{Te}_5$ (GST, crystal phase, $n_c = 4.01 + i3.61$)/ $\text{ZnS-SiO}_2/\text{air}$ ($n = 1$). The figure shows that the layer structure between the insulating layer (the second ZnS-SiO_2) and the record mark (GST) is deformed after the writing process. The nanosize Ag nanoparticles inside the bubble pits are embedded in the AgO_x layer and are assumed to move under the influence of the readout laser beam, as shown by previous experimental work.¹⁶⁾ The dispersive behavior of Ag and GST is simulated by the Lorentz model.¹⁷⁾ The AgO_x layer is sandwiched between the insulating ZnS-SiO_2 layers and a GST-alloy phase-change data layer is located directly above this trilayer structure. The other parameters that were used were polarization [transverse magnetic (TM)-polarization], cell size ($\Delta x = \Delta y = 0.5 \text{ nm}$), and the refractive index of Ag ($n = 0.055 + i4.44$). The incident Gaussian beams full width at half-maximum (FWHM) was adjusted such that $\text{FWHM} = 0.51\lambda/\text{NA}$.¹⁸⁾ The phase-change data layer is in a on-mark situation. The on-mark situation means that the incident light focuses on the recording mark. The numerical aperture (NA) of the objective lens is 0.85.

As studied by our research group,^{19,20)} the density of the silver nanoparticles can be controlled by increasing or decreasing the incident laser power in the photodissociation process. A lower nanoparticles density shows less interaction between the nanoparticles. If the density of the nanoparticles is increased, the local fields are coupled together and produce more evanescent fields from adjacent nanoparticles. Figures 2(a) and 2(b) show the near-field distributions of Ag nanoparticles of deformed and normal (undeformed) AgO_x -type Super RENS structures, respectively. The total number of nanoparticles is 82 in the deformed type and 57 in the

normal type, each with a diameter of 4 nm. The wavelength of incident light is $\lambda = 650 \text{ nm}$ for TM polarization. In Figs. 2(a) and 2(b), highly localized enhancements and interactions are produced between adjacent Ag nanoparticles in the near-field zone. When the free electrons of an Ag nanoparticle oscillate collectively in resonance with the incident light, the interaction may constitute a surface plasmon polariton (SPP). The field enhancement is due to SPPs. Each embedded Ag nanoparticle behaves like a dipole and a very strong scattering center. The local fields from adjacent nanoparticles are coupled together and produce highly enhanced evanescent fields. Since the boundary conditions of Maxwell's equations stipulate that induced surface charge density is proportional to the discontinuity of the electric field component normal to the surface, only TM-polarized light can excite SPPs. These near-field optical signals of the Ag nanoparticles correspond to the nonlinear near-field optical properties previously found in experiments.²¹⁾

The Ag nanoparticles inside the bubble pits are assumed to be under the influence of a readout incident beam. The donut shape aperture (deformed AgO_x film) is formed only in the on-mark situation of the readout beam and provides the additional length (or path) longer than the normal length. In comparison with the evanescent fields for the normal type [Fig. 2(b)], the bubble pits of the deformed type [Fig. 2(a)] give additional outer boundaries to the motion of the Ag nanoparticles, and excite more evanescent fields, which are located at the far edge (top surface) of the bubble from the optical axis of the incident beam. It can be clearly seen in Fig. 1(a) that more evanescent fields extend in the direction of the y -axis (propagation direction). This indicates that the bubble and particles tend to increase the readout signal intensity.

From experimental observation, the exact shape of a Ag nanoparticle is between a circle and a square. When the diameter of a nanoparticle is increased from 3 to 7 nm, the shape becomes closer to a circle. The optical responses of randomly distributed Ag nanoparticles are complicated, however, one key method for understanding these effects is to distinguish the contributions from each nanoparticle and particle-particle interactions. SPPs and the scattering efficiency of Ag nanoparticles are usually sensitive to and controllable by particle size, the surrounding medium and the wavelength of the incident light. With the same distribution of Ag nanoparticles as shown in the Fig. 1, the field intensity measured at the surface of the deformed AgO_x layer varied considerably when they were illuminated by incident light at various frequencies (wavelengths). To verify the effect of Ag nano particle size, the variation in near-field intensity measured at the top surface of the deformed AgO_x film with increasing diameter is tested. Figure 3 shows field intensity as a function of the diameter of Ag nanoparticles from 1 to 7 nm, in increments of 1 nm, measured at the top surface of the deformed AgO_x film at various wavelengths of incident light. We chose the wavelengths of incident light of 350, 450, 550, 650, and 750 nm, respectively, as examples, and the surface plasmon effects of other wavelengths can be verified by the same procedures. In the finite area of the deformed AgO_x layer and the fixed total number of 82 Ag nanoparticles shown in Fig. 1, a smaller

Fig. 2. Calculated results of near-field distributions of Ag nanoparticles in deformed and normal (undeformed) AgO_x -type Super-RENS structure, respectively. The total number of Ag nanoparticles is 82 in the deformed type and 57 in the normal type, each with a diameter of 4 nm. The wavelength of incident light is $\lambda = 650 \text{ nm}$ and the NA of the lens is 0.85 for TM polarization.

Fig. 3. Calculated results of field intensity as a function of diameter of Ag nanoparticle from 1 to 7 nm, in increments of 1 nm, measured at the top surface of the deformed AgO_x film at various wavelengths of incident light. We chose the wavelengths of incident light of 350, 450, 550, 650, 750 nm, respectively.

size of Ag nanoparticles means a lower density because the gap between two particles is larger. In the same manner, a larger size means a higher density cases because the gap between two particles is smaller.

The results in Fig. 3 clearly show that the size variations of Ag nanoparticles may yield different field intensities and indicate that the intensities of local fields exhibit a nonlinear optical response with increasing nanoparticle diameter. When particle diameter is less than 3 nm (the lower density cases), shorter wavelengths (e.g., $\lambda = 350$ and 450 nm) can excite more evanescent fields from the surface of Ag nanoparticles than longer wavelengths (e.g., $\lambda = 550$, 650 , and 750 nm). When particle size is larger than 3 nm (the higher density cases), the gaps between Ag nanoparticles are smaller and more local fields from adjacent nanoparticles are coupled together to produce highly enhanced evanescent fields, for longer wavelengths. It can be seen in Fig. 3 that the field intensity at a fixed particle diameter of 6 nm exhibited peak values at the longer wavelengths (e.g., $\lambda = 550$, 650 , and 750 nm), and these were the points where surface plasmon resonance occurred. This indicates that, at these wavelengths, the particle diameter of 6 nm provides a major contribution to plasmon coupling with the dipole radiation between the particles being the evanescent field of near-field photons. Since the field propagation is predominantly taken as the near-field photons, super-resolution exceeding the diffraction limit is made similar to that of near-field scanning optical microscopy. When the particle size is increased to 7 nm, the Ag nanoparticles form aggregative silver clusters which act as a silver bulk. The intensity decreases abruptly owing to absorption in the metal bulk and the destructive interference of SPPs in particle-to-particle interactions.

In further research, we will consider the model where not a single bubble pit but two or more pits are in one spot. However, owing to the varying power of incident light, the distribution and density of the nanoparticles may not be uniformly random in the area of the realistic deformed AgO_x layer. Further research on the effects of different distribution patterns may yield a better understanding of the optical

properties of the deformed AgO_x -type Super-RENS. Moreover, on the basis of results of this study, we will calculate the change in intensity (difference signal) in the far-field by varying mark size, the distribution, and the number of metallic particles in the active layer that become detectable signals.

In conclusion, the deformation and plasmon effects of collective localized surface plasmons in a deformed AgO_x -type super-RENS structure have been studied using two-dimensional FDTD simulations. We find that polarization direction, the wavelength of incident light, and the size of Ag nanoparticles are sensitive to the plasma resonance. The Ag nanoparticles inside the deformed AgO_x -type super-RENS structure provide an additional length (or path) that is longer than the normal length, give additional outer boundaries to the motion of the Ag nanoparticles, and excite more evanescent fields that are located in the far edge of the bubble pit from the optical axis of the incident beam. The above research can help us to design a super-RENS with superior resolution, as well as an alternative approach to near-field optical research for plasmon device, biological sensors and nanophotonic devices.

The authors gratefully acknowledge the reviewer's helpful suggestions and thank Ying Zhou for comments on simulation structure. We are thankful for the financial support from National Science Council, Taiwan, R.O.C., under grant number NSC 94-2112-M-231-001, and the Ministry of Economic Affairs, Taiwan, R.O.C., under grant number 94-EC-17-A-08-S1-0006.

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