Imaging Properties of Three Dimensional Aperture Near-Field Scanning Optical Microscopy and Optimized Near-Field Fiber Probe Designs

Yuan-Fong CHAU, Tzong-Jer YANG* and Din-Ping TSAI¹

Department of Electrophysics, National Chiao Tung University, Hsinchu, Taiwan, Republic of China

¹*Department of Physics, National Taiwan University, Taipei, Taiwan, Republic of China*

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A series of finite difference time domain (FDTD) methods for obtaining more insight on the near-field distribution of subwavelength aperture and fiber probes are numerically investigated. The treatment of dispersive materials in a time-domain-dependent fashion is considered. Several significant factors are also considered successively, *e.g.*, the near-field distribution of subwavelength aperture in an infinite aluminum plane with sample interactions, the characteristics between the near-field distribution and the depolarization phenomenon, different types of three-dimensional tip (noncoated and metal-coated) used to illuminate a photosensitive sample and the polarization of an incident electromagnetic field. Moreover, the FDTD designs of two types of improved probe are illustrated and a suggestion for fabricating an optimal probe is given. Our proposed structures will yield useful information and guidelines for designing high-performance near-field probes.

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1. Introduction

The major problem in near-field scanning optical microscopy (NSOM)^{1–4)} is the determination of field distributions and radiation properties of a given probe/sample structure. Therefore, the precise spatial resolution depends on such factors as the taper angle, diameter of tip opening, refractive index of inner dielectric material of coated probes, thickness and shape of metal coating, and polarization and mode patterns of excitation, all of which can affect the field pattern near the tip. In addition, the presence of the object being scanned can affect the near-field pattern, depending on the electromagnetic properties of the object, causing the field pattern to change from its unobstructed behavior. A better understanding of field pattern will lead to an unambiguous analysis of an NSOM image and will make NSOM a more reliable imaging method for both biologically interesting samples and nanostructured materials. Due to the complex boundary conditions imposed by numerical methods only, requiring an extended computational work, experimental limitations in general prevent the direct probing of near fields. Thus, the questions of achievable resolution and contrast have been left mainly to heuristic arguments and experimental evidence. This also requires a correct interpretation of NSOM images, which is a trivial task as will be seen from the results to be presented here. In the past decade, several efficient methods were implemented to overcome the difficulties raised by the complex geometry of such optical systems. Memory requirement is important in particular for calculating a three-dimensionally distributed field with a probe and sample. It has been carried out by the dipole approximation method (DAM),⁵⁾ the boundary element method (BEM),⁶⁾ the multiple multipole method (MMP),^{7,8)} the finite element method (FEM),⁹⁾ the Green tensor method,^{10,11)} and the FDTD method.^{12–14)} Among these methods, the FDTD method reduces the computer memory amount of the same 3D model and solves Maxwell's equations without any simple approximations other than the discretized grid. This method is well suited in

simulating near-field configurations and has been proved to be fruitful in solving near-field optical problems.^{15–21)} The resolution provided by the two-dimensional (2D) model NSOM is lower than that expected on the basis of experiments and cannot handle a complex probe-sample system. Of the several possible reasons, the soft decay of the fields in 2D ($r^{-1/2}$ instead of r^{-1}) might be the most fundamental reason. This shows the need for 3D calculations which, however, require much computation times. Treating metals as perfect conductors^{22–24)} causes concern that the optics in the near-field region surrounded by the metals may not be simulated accurately. In this paper, the treatment of dispersive metals in a time-domain-dependent fashion is considered. Little has been known if and how the emerging light from the tip loses the characteristics of polarization after exiting from the tip apex and if produces two perpendicularly polarized electric field components. This near-field effect phenomenon will be described here.

In this study, we investigate the near-field mechanism between a tip and a sample using a three-dimensional (3D) model and the different aspects of this local interaction. First, we will discuss some preliminary results recorded with the near-field distribution of subwavelength circular aperture with sample interactions on an infinite aluminum thin plate to construct the foundation of latter analysis. Next, in the following section, we will compare the field distributions produced by two types of tip (noncoated and metal-coated) and give a suggestion for fabricating an optimal tip.

2. Numerical Method

The finite difference time domain method is a flexible numerical means of solving electromagnetic problems by integrating Maxwell differential equations,

$$\nabla \times \mathbf{H} = \varepsilon(\mathbf{r}) \frac{\partial \mathbf{E}}{\partial t} + \sigma(\mathbf{r}) \mathbf{E}, \quad \nabla \times \mathbf{E} = -\mu(\mathbf{r}) \frac{\partial \mathbf{H}}{\partial t}, \quad (2.1)$$

for an arbitrary geometry in three-dimensional space, where $\varepsilon(\mathbf{r})$, $\mu(\mathbf{r})$, and $\sigma(\mathbf{r})$ are the position-dependent permittivity, permeability, and conductivity of the material, respectively. The vectorial wave equation, given by these equations, must then be solved under the specific conditions imposed by the

*E-mail address: yangtj@cc.nctu.edu.tw

nature and the geometry of the tip-sample junction. When the surfaces limiting the illuminated objects display a complex shape simultaneously with strong variations of the dielectric functions, seeking solutions for these universal equation requires specific care. We employed a numerical technique known as the finite difference time domain (FDTD) method for this analysis. The FDTD method treats Maxwell equations as a set of finite difference equations in both time and space. The model space considered includes both the probe and the sample surface and consists of cubic cell aggregates. Each cell aggregate divides the model to be analyzed into a gridwork of small square or cuboidal cells, called "Yee cells". Each Yee cell is positioned as a vector component of the electric and magnetic fields that gives the cell location. This object definition mechanism is the heart of the flexibility of the FDTD method, as any arbitrary geometry can be simulated. Each cell made susceptible to a given material. The FDTD method calculates the electric and magnetic fields in each cell by integrating Maxwell equations in a "leap-frog" fashion until the steady state is reached.¹⁴⁾ In the case of Gaussian pulse modulating with a sinusoidal excitation source, the steady state is reached (that is, the fields are not changing in the peak-to-peak amplitude) when all scattered fields vary sinusoidally in time. The size of each cell is limited on the upper bound to be no longer than about one-tenth of the wavelength in order to assure a sufficient sampling of the spatially varying fields. However, near-field problems are well within this length limitation, and in fact, there is no lower limit on the size of the Yee cell in terms of wavelength. Such small cells result in extremely short time steps in the simulation, as the FDTD algorithm is stable only if

$$\Delta t \leq \frac{1}{c\sqrt{\Delta x^{-2} + \Delta y^{-2} + \Delta z^{-2}}}, \quad (2.2)$$

where c is the speed of light and Δ is the side of a cubical cell. This is a result of the Courant stability condition, which is based on the fact that the plane wave traveling in FDTD space does not travel through more than one cell in space and time.

The finite difference equations corresponding to Ampere's law based on a central difference approximation result in an explicit time-stepping algorithm, whose finite difference form for the z -component of electric field in any given Yee cell can be written as

$$\begin{aligned} E_z^n\left(i, j, k + \frac{1}{2}\right) = & \frac{1 - \frac{\sigma(i, j, k + \frac{1}{2})\Delta t}{2\varepsilon(i, j, k + \frac{1}{2})}}{1 + \frac{\sigma(i, j, k + \frac{1}{2})\Delta t}{2\varepsilon(i, j, k + \frac{1}{2})}} E_z^{n-1}\left(i, j, k + \frac{1}{2}\right) \\ & + \frac{\Delta t}{1 + \frac{\sigma(i, j, k + \frac{1}{2})\Delta t}{2\varepsilon(i, j, k + \frac{1}{2})}} \left[H_y^{n-\frac{1}{2}}\left(i + \frac{1}{2}, j, k + \frac{1}{2}\right) \right. \\ & - H_y^{n-\frac{1}{2}}\left(i - \frac{1}{2}, j, k + \frac{1}{2}\right) + H_x^{n-\frac{1}{2}}\left(i, j - \frac{1}{2}, k + \frac{1}{2}\right) \\ & \left. - H_x^{n-\frac{1}{2}}\left(i, j + \frac{1}{2}, k + \frac{1}{2}\right) \right]. \end{aligned} \quad (2.3)$$

The magnetic induction elements H_x are calculated using Faraday's law of induction:

$$\begin{aligned} H_z^{n+\frac{1}{2}}\left(i + \frac{1}{2}, j + \frac{1}{2}, k\right) = & H_z^{n-\frac{1}{2}}\left(i + \frac{1}{2}, j + \frac{1}{2}, k\right) \\ & - \frac{\Delta t}{\mu(i + \frac{1}{2}, j + \frac{1}{2}, k)\Delta} \left[E_y^n\left(i + 1, j + \frac{1}{2}, k\right) \right. \\ & - E_y^n\left(i, j + \frac{1}{2}, k\right) + E_x^n\left(i + \frac{1}{2}, j, k\right) \\ & \left. - E_x^n\left(i + \frac{1}{2}, j + 1, k\right) \right]. \end{aligned} \quad (2.4)$$

The updated equations for x - and y -components are obtained by rotation of indices, where the subscript on each field gives its polarization and cell location. $\Delta x = \Delta y = \Delta z = \Delta$ are the lengths of the cell in three directions, respectively, Δt is the unit time increment, the superscript n is for time step, the refractive index is related to the permittivity ε by $\sqrt{\varepsilon}$, $(i\Delta x, j\Delta y, k\Delta z)$ is a grid point, $\varepsilon(i, j, k)$ is the permittivity at $(i\Delta x, j\Delta y, k\Delta z)$, and similarly, electrical conductivity $\sigma(i, j, k)$ and permeability $\mu(i, j, k)$.

Special consideration should be given at the boundary of the finite computational domain, where the fields are updated using special boundary conditions, as information outside of the domain is not available. The outside of the simulation area is considered to be the boundary of perfectly matched layer (PML) absorption;²⁵⁾ thus the electromagnetic field arriving at the boundary is not reflected but is perfectly absorbed by the PML. Hence, one can determine the electromagnetic field inside the calculated space.

Typically, FDTD calculations have been carried out for dielectric materials. Noble metals (aluminum, gold, silver, and copper) in the optical regime possess complex refractive indices whose imaginary part is greater than their real part. At optical frequencies, the permittivity ε , which is defined in the frequency domain, is complex, with the real part of the permittivity being negative.^{26,27)} Inserting a negative real permittivity in the FDTD algorithms causes unstable situations in a simulation, which results in an apparent sign change in $D(\omega)$ (the Fourier transform of electric displacement). The sign change is caused by a material response that is 180° out of phase with respect to the electric field incident on the medium. To represent real metals correctly and maintain numerical stability require that a second-order material model for the medium be included in the FDTD scheme. The complex refractive indices of many materials including the noble metals have been measured and tabulated in the *CRC Laser Handbook*.²⁸⁾ The materials with frequency-dependent complex dielectric constants have been developed for treating dispersive materials in a time-domain-dependent fashion.²⁹⁾ The relation among electric displacement D , dielectric constant ε , and electric field E is

$$D(r, t) = \int_0^t \varepsilon(r, \tau) E(r, t - \tau) d\tau. \quad (2.5)$$

We used the recursive convolution scheme (RC) method to evaluate the system which includes a dispersive medium such as a metal films.³⁰⁾ The relative dielectric constant in the frequency domain is

$$\varepsilon_r(\omega) = \varepsilon_\infty + \chi(\omega), \quad (2.6)$$

where ε_∞ is the infinite frequency permittivity, χ is the susceptibility, and ω is the angular frequency. If $\chi(\infty) = 0$, eq. (2.5) can be expressed as

$$\mathbf{D}(t) = \varepsilon_0 \varepsilon_\infty \mathbf{E}(t) + \int_0^t \chi(\tau) \mathbf{E}(t - \tau) d\tau. \quad (2.7)$$

Here, $\chi(\tau)$ is the Fourier-transformed $\chi(\omega)$ and ε_0 is the permittivity of free space. The frequency domain Lorentz dispersion model can be designed to give the correct refractive index for any material at a single ω .

The refractive index associated with the Lorentz model, $n_L(\omega)$, has the form

$$n_L^2(\omega) = \varepsilon_R(\omega) = \frac{\chi_0 \omega_0^2}{\omega_0^2 - \omega^2 + j\Gamma\omega} + \frac{\varepsilon_\infty}{\varepsilon_0}. \quad (2.8)$$

where ε_R , ω_0 and Γ are the total complex permittivity, the resonant frequency and the damping coefficient for the model, respectively. The relations between ε_R and n_r (the real part of the refractive index) and n_i (the imaginary part of the refractive index) can be expressed as

$$\begin{aligned} \varepsilon_R(\omega) &= (n_L(\omega))^2 = (n_r^2 - n_i^2) + 2jn_r n_i, \\ \varepsilon(\omega) &= 1 + \frac{\omega_p^2}{\omega(\omega_c - \omega)} = \varepsilon_\infty + \chi(\omega), \end{aligned} \quad (2.9)$$

where ω_p is the radian plasma frequency and ω_c is the collision angular frequency. The Drude model reveals a simple physical picture of a free electron gas.³¹⁾ The response of near-field probes has been simulated using the Drude model with the FDTD method with some success.²¹⁾ We note that this Lorentz dispersion model contains the standard Drude model for metals as a limiting case. One obtains the Drude model by taking the limit $\omega \gg \omega_0$ and introducing the plasma frequency $\omega_p^2 = \chi_0 \omega_0^2$ and the collision frequency $\omega_c = \Gamma/2\pi$. For calculating the permittivity of the materials, one can choose a reasonable DC and high-frequency response χ_0 and ε_∞ , and then solve for ω_0 and Γ .

$$\omega_0^2 = \frac{\omega^2}{1 - \chi_0(\varepsilon_r - \varepsilon_\infty/\varepsilon_0)/[(\varepsilon_r - \varepsilon_\infty/\varepsilon_0)^2 + \varepsilon_i^2]}, \quad (2.10)$$

$$\Gamma = \frac{\omega \chi_0 \varepsilon_i}{(\varepsilon_r - \varepsilon_\infty/\varepsilon_0)^2 + \varepsilon_i^2 - \chi_0(\varepsilon_r - \varepsilon_\infty/\varepsilon_0)}, \quad (2.11)$$

The parameters are adjusted to closely match the experimentally observed optical properties. We set $\chi_0 = 10$ and $\varepsilon_\infty = 2.98 \varepsilon_0$ in this work. The refractive index of the metal material (aluminum) in our simulations at wavelength $\lambda = 633 \text{ nm}$ ($\omega = 1.71 \times 10^{14} \text{ s}^{-1}$)³²⁾ is $-34.5 + i8.5$.^{33,34)} From eqs. (2.10) and (2.11), ω_0 and Γ are obtained for the material and the time domain version of the Lorentz model with these parameters is included in the FDTD computation in the metal film region of the numerical mesh.

3. Models and Results

3.1 Subwavelength circular aperture image with sample interactions

The optical properties of metals have been of interest for more than 100 years.³⁵⁾ In the past century, there were many important studies about it, such as that on a solution to the problem of light scattering by spherical metal particles.³⁶⁾ Other high-symmetry materials, including finite-thickness

slabs, cylinders, and ellipsoids, have been analytically solved.³⁷⁾ The analytical solutions require only the particle size and frequency-dependent index of refraction $n = n_r + jn_i$, with the real part, n_r , representing the index of refraction, and the imaginary part, n_i , representing the extinction coefficient. In the real near-field optical recording systems, the light can be expressed as a wave vector and consists of only a real component in the normal state. The wave vector has a scale value $2\pi n/\lambda$, where n and λ are the refractive index and wavelength, respectively. In metals, the component of the wave vector will be larger than the above scalar value if it is dispersed from the area of high frequency or total internal reflection from the interface between two different materials. The component of the wave vector becomes purely imaginary, and thus this wave cannot be a propagation wave but an evanescent wave in one spatial direction, resulting in an exponential decay within a small distance compared to its wavelength. Since the optical properties in near-field zone are much different from those in the far-field zone. However, we can obtain a high-quality resolution beyond the diffraction limit of an optical wave if we use the evanescent wave within a very short distance from the interface. The resolution of the evanescent wave is not related to its wavelength. It reaches below 100 nm and has already been applied in NSOM.³⁸⁾ Although the thin metal plate with a subwavelength circular aperture with sample interactions is a simple case, we think it is important to the near-field region and necessary for a more detailed analysis of the foundation of our latter simulations.

The two sets of equations described by eqs. (2.3) and (2.4) are alternately evaluated to simulate the light propagation. In the past decade, two different setups were developed from this basic configuration: (1) the transmission illumination mode in which the light radiated by a nanometric emitter is converted into propagating waves by the sample itself and (2) the transmission collection mode which is easily obtained by reversing the light path. The aperture serves as a nanocollector, and the light converted in the near-field zone is transmitted to a photodetector. We will discuss the transmission collection mode in this section. Figure 1 shows

Fig. 1. (a) Sectional plane diagram of 3D subwavelength circular aperture with sample interactions. (b) Convention used when considering the effects of two orthogonal polarization states (s- and p-polarizations). The incident electric field is p-polarized when the electric field vector oscillates in a sectional plane that is perpendicular to the surface of the edge structure. On the other hand, s-polarization occurs when the field vector oscillates in the normal direction to a sectional plane.

Fig. 2. Distribution of the 3D electric field intensity modulus around the aperture-sample coupling zone: (a) in x - z plane (at $y = 50\Delta$), p-polarization; (b) in y - z plane (at $x = 50\Delta$), p-polarization; (c) in x - z plane (at $y = 50\Delta$), s-polarization; and (d) in y - z plane (at $x = 50\Delta$), s-polarization. The electric field results from an incident wave coming from the left side. In the gray scale used, white is used to signify high intensities.

a schematic diagram of the 3D subwavelength circular aperture with a sample model to be analyzed. It has an opening of 80 nm (40 cells) in an infinite aluminum thin metal plate with a thickness of 40 nm (20 cells) and a dielectric sample with a refractive index of $n = 1.5$. The distance between the aperture and the sample surface is 20 nm (10 cells); the aperture at the bottom of the model is filled with silica core, as shown in Fig. 1. The surface profile of the dielectric sample consists of a flat surface with a single cylindrical protrusion whose height and diameter are 20 nm (10 cells) and 120 nm (60 cells), respectively. Another difficulty in the FDTD method is the error caused by the incident light on the surface. A total field/scattered field formulation¹³⁾ is used to introduce the incident light into the calculation space. The frequency of the source is 4.74×10^{14} Hz, corresponding to a free-space wavelength of 633 nm, which propagates from the base plane to the aperture, located in the plane shown in Fig. 1. The dimensions of each cell are $\Delta x = \Delta y = \Delta z = \Delta = 2$ nm, and the total space volume considered consists of $100(x) \times 100(y) \times 100(z)$ cells.

The effects of light polarization on the generated near-field images are studied under both parallel and perpendicular polarization conditions. When the model structure possesses a preferred orientation, the characteristics of the scattered light depend on the polarization direction (either parallel or perpendicular) relative to the structure orientation. In this paper, we report that the incident electric field is p-polarized when the electric field vector oscillates in a sectional plane that is perpendicular to the surface of the edge structure. On the other hand, s-polarization occurs

when the field vector oscillates in the normal direction to a sectional plane.

Figures 2(a)–2(d) show the calculated results of the three-dimensional electric field for p-polarization and s-polarization illuminations. In the gray scale used, white is used to signify high intensities. Although this model is a rotational symmetric system, the electric field distributions formed in the two orthogonal cross sections are different from each other due to the difference between the boundary conditions at the edge interface. This result implies that the resolution of an aperture NSOM is given by the size of the aperture rim, which is a great advantage of an aperture NSOM. In an NSOM with an aperture in a metal coating,¹⁵⁾ the penetration of photons through the metal coating smears the spot; thus the practical minimum diameter of the spot is at least 50 nm even if an extremely small aperture is used. In the sectional image of the model illuminated by p-polarization shown in Figs. 2(a) and 2(b), the intensity becomes stronger near the edges of the aperture and sample, and the image generated agrees reasonably with the geometrical profile of the same model shown in Fig. 1. The field enhancement is due to the surface plasmon polariton because the thin-film metal coating is excited at the aperture rim by the evanescent photons and propagates nearly parallel to the sample surface, resulting in the enlargement of the spot. Since the electric flux density is conserved, the electric field amplitude is greater than the external medium with a smaller permittivity.

Considering the boundary condition, the incident wave should have a component vertical to the subwavelength circular aperture in an infinite aluminum thin plate at the output edge of the aperture in order to excite the localized-

mode surface plasmon.³⁹⁾ Figures 2(c) and 2(d) show the same results as in Fig. 2(a) and 2(b), except that the illumination is s-polarized. In the central part of this model, the electric intensity is stronger than those in other regions of the model. Moreover, it is found that the images contrast decreases relative to the images shown in Figs. 2(a) and 2(b). In this case, s-polarization is responsible for surface digging. The estimated digging of the surface after illumination by a metallized aperture with s-polarization is similar to the experimental surface deformation, which is in good agreement with current experimental data. The main differences between the two principal polarizations are hence the degree of confinement which is favorable for s-polarization and the available electric field distribution which is favorable for p-polarization. It is important to note here that the intensity of p-polarized field plays a leading role in edge enhancement. If we observe again the field maps in Figs. 2(a)–2(d), we can see that the light is generated mostly by the aperture rim in p-polarization, due to the excitation of the metal coating and the presence of a local dipole along the aperture rim. In contrast, for s-polarization, large amount of light at the central part of the aperture is responsible for surface digging.

3.2 3D NSOM image of dielectric tip

The perspective of improving the optical data storage is a big challenge at the beginning of the communication century. Different methods were then investigated to achieve this aim, at various efficiencies. The illumination probe can be a dielectric probe,⁴⁾ a coated tip,⁴⁰⁾ or even an STM tip.⁴¹⁾ Possible photosensitive samples are numerous, from the magneto-optical sample⁴²⁾ to the liquid-crystal layer.⁴³⁾ Various parameters are involved and a lot of progress has still to be realized to control them. We will compare a noncoated (purely dielectric) tip with a metal-coated tip here. The two tips can also be used as optical pickups for a data storage system to realize a very high density data recording. The performance of the NSOM or data storage system depends on the optical properties of the near-field tip. Both high throughput (light efficiency) and resolution (smaller spot size) are desired. The transmission-illumination-mode NSOM allows the radiative optical wave to be converted into a confined wave concentrated near the probe apex.

Figures 3(a) and 3(b) show a sectional plane diagram of the 3D NSOM model of the dielectric tip and metal-coated

tip, respectively, in which probe-sample interactions are taken into account in order to realize the field distribution formed by the probe tip. In Fig. 3(a), the dielectric probe tip exhibits a cone profile and has a height of 400 nm and an aperture openings of 50 nm (10 cells) at the apex and 400 nm (80 cells) at the bottom of the probe. We choose to model an acute tip with a half-angle slope of 23.6° . The material of the probe is purely dielectric with a refractive index of 1.83, except for a thin (50 nm) metal film screen at its bottom surface that is assumed to center the incident beam to the axis of the tip. In front of the probe, at a distance of 30 nm (6 cells), a semi-infinite silicon medium was taken as the sample with a refractive index of $n = 1.5$. The dimensions of each cell are $\Delta x = \Delta y = \Delta z = \Delta = 5$ nm, and the entire region model consists of $140(x) \times 140(y) \times 140(z)$ cells. In the experiments, the three-dimensional probes support both polarizations simultaneously. The output field distribution of the tip will be small as the tip size decreases. Thus, it is expected that the optical power density of the field distribution is maximum at a certain area of the tip due to the focusing effect by the reflection on the internal surface of the cone-shaped tip. Figures 4(a)–4(d) show the calculated results in the x - z sectional (at $y = 70\Delta$) and y - z sectional (at $x = 70\Delta$) plane for p-polarization and s-polarization illuminations. These results show good agreement with the experimental report by Danzebrink *et al.*⁴⁴⁾ As expected, the field intensity decreases rapidly near the probe tip due to the conversion of the light wave to the radiation mode or its reflection. That is, the incident wave from the base plane to the tip propagates inside the tip as long as the cone-shaped tip is sufficiently wide. Once the propagation becomes too difficult, more light escapes from the lateral surfaces, thus limiting the tip. These far-field components will contribute to illuminate a large spatial zone. The light generated by the apex of the tip is essentially evanescent, since most propagating components escaped laterally before reaching the coupling zone. The near-field effect is confined to the surface area in front of the tip extremity. In this region, the illumination process is mainly supported by evanescent fields. The distribution of the field intensity along the surface is similar to that obtained when an incident field is diffracted by a subwavelength aperture. On the basis of the results of our simulation in this case, we find the electric field leaking from the conic side much stronger than that emerging from the tip apex for noncoated (purely dielectric) probes. The reflected light from the conic surface inside the probe couples with the incident light to form an incomplete standing wave and escapes from the lateral surfaces as long as the diameter of the cone-shaped tip is approximately less than 180 nm. This wave will form the far-field component.

The depolarization phenomenon of electric field components is important in the near-field zone. The dielectric tip decreases the intensity of the polarized incident wave, namely, the emerging light from the tip loses the characteristics of polarization, after exiting from the tip apex and produces two perpendicularly polarized electric-field components; that is, the incident field is entirely polarized along the y -axis and the scattered field is divided into the components of the electric fields along the x -axis and z -axis. These two components produce depolarization at the interface between the probe tip and the air. Figure 5 shows

Fig. 3. Sectional plane diagram of 3D NSOM model for (a) a dielectric tip and (b) a metal-coated tip, in which probe-sample interactions are taken into account.

Fig. 4. Distribution of the 3D electric field intensity modulus around the tip-sample coupling zone (dielectric tip): (a) in x - z plane (at $y = 70\Delta$), p-polarization; (b) in y - z plane (at $x = 70\Delta$), p-polarization; (c) in x - z plane (at $y = 70\Delta$), s-polarization; and (d) in y - z plane (at $x = 70\Delta$), s-polarization. The electric field originates from an incident wave coming from the left side.

Fig. 5. Distributions of total electric field and field components without sample interactions for p-polarization illumination in the plane away from probe apex of $z = 5$ nm, 15 nm, 30 nm (from left to right). From top to bottom: $|E_t|$, $4 \times |E_x|$, $|E_y|$, and $|E_z|$. The size of each image plane displayed is $700 \times 700 \text{ nm}^2$ in front of the probe.

Fig. 6. Distribution of the 3D electric field intensity modulus around the tip-sample coupling zone (metal-coated tip): (a) in x - z plane (at $y = 70\Delta$), p-polarization; (b) in y - z plane (at $x = 70\Delta$), p-polarization; (c) in x - z plane (at $y = 70\Delta$), s-polarization; and (d) in y - z plane (at $x = 70\Delta$), s-polarization. The electric field originates from an incident wave coming from the left side.

the distributions of total electric field without sample interactions in the x - y sectional plane for p-polarization illumination and the field components in the plane away from the probe apex of $z = 5$ nm, 15 nm, and 30 nm (from left to right). From top to bottom: $|E_t|$, $4 \times |E_x|$, and $|E_y|$, $|E_z|$. The size of each image plane displayed is $700 \times 700 \text{ nm}^2$ ($140(x) \times 140(y)$ cells) in front of the probe. The x -component of electric fields forms a four-petal field pattern distributed symmetrically along the tip apex rim, which is much smaller than the y - and z -components of electric fields. The z -component of electric fields forms a two-petal field pattern distributed symmetrically along the tip apex rim, which is also smaller than the y -component of electric fields; a marked enhancement occurs at the tip apex rim. Both $|E_x|$ and $|E_z|$ decay rapidly as the distance away from the probe increases. The y -component of electric fields induces propagation mainly in the forward direction along the optical axis and possesses the total field distribution mostly emerging from the tip.

The depolarization phenomenon of the components of electric fields is the near-field effect. Recall that using the weak scattering probe (Rayleigh particle), where the interaction of the tip with the electromagnetic field near the sample surface is approximately negligible, the strongest intensities are found near the center of the tip apex. As shown in Figs. 4(c) and 4(d), obviously, for this noncoated probe, s-polarization is responsible for surface digging, while p-polarization generates a large-scale background in which a central depression, due to the near-field couplings, is superimposed.

3.3 3D NSOM image of metal-coated tip

For the metal-coated tip, the model is used in the same manner as in the noncoated dielectric tip except that it is coated with an aluminum film of 25 nm thickness (5 cells) and its length is 350 nm (70 cells) as shown in Fig. 3(b). Figures 6(a)–6(d) show the calculated results in the x - z sectional plane (at $y = 70\Delta$) and y - z sectional plane (at $x = 70\Delta$) for p-polarization and s-polarization illuminations, respectively. The presence of a metal coating on the tip prevents the lateral escape of light and significantly reduces the penetration of light emerging from the probe. The reflection light from the metallic layer of the cone-shaped tip couples with the incident light to form a high-intensity standing wave and becomes maximum at the diameter of the sectional plane in cone-shaped tip of about 320 nm. Note that this diameter is different from that of the noncoated tip (180 nm).

Metals are good materials for the confinement of the light wave in the tip since they have high absorption properties due to their high values of the imaginary part and negative dielectric constants. We choose Al as the coating film on the probe surface. In Figs. 6(c) and 6(d), the incident wave is s-polarized, and penetrates inside the tip without escaping through the lateral surfaces. It propagates mostly in the forward direction forward the apex, with an intense field emerging from the aperture. However, the field intensity produced by the tip is weaker than that produced by the noncoated tip. On the other hand, when considering the p-polarization illumination in Figs. 6(a) and 6(b), the field intensity at the tip apex is not weak. The higher field intensity near the tip aperture rim originates from the

evanescent components that were stopped before reaching the tip end. Note the localized electric field enhancement at the edge along the metal tip rim extending in several ten nanometer range from the tip apex. The field enhancement of a metallic tip originates mainly from the localized surface plasmon mode excited at the apex of the tip by the evanescent field.

In addition, when the thickness of the coating of a probe is very small, such as less than 100 nm, the leakage of incident light deteriorates the near-field signal in an experiment. The reasons why we still used a thinner film (25 nm) coating are as follows: (1) In our study, we are interested in the surface plasma around the tip aperture rim. Plasmons may play a role in the interaction of optical radiation with an aperture. In the near-field zone, the thinner the metal-coated probe, the higher the magnitudes of the effects of surface atoms. Indeed, according to the frequency-dependent dielectric properties, localized eigenmodes characterized by evanescent wavefunctions may be sustained by small objects and even by surfaces. If a thinner metal thin film is coated along the tip apex rim, the aperture of the tip can be regarded as many dipoles at the symmetry positions around the circular aperture. The metal-coated tip we used in Fig. 3(b) has an aperture opening of 50 nm at its apex. If we choose a coating thicker than 100 nm for the probe, the effect of the near field will not be obvious as compared to a thinner one. (2) In order to gain a better understanding of plasmon excitation in the near-field region, a shorter distance between the tip and sample (30 nm in this section), a smaller diameter of the tip apex (50 nm in this paper), and a thinner metal coating (25 nm in this paper) are needed in the simulation domain. (3) On the basis of the results of our simulations, we find that metal coatings of different thicknesses possess different cutoff planes inside the probe tip. These so-called cutoff planes are distinct planes between the radiation waves and evanescent fields. The thicker the coating, the shorter the cutoff plane near the bottom surface of the probe. This is due to the difference in boundary condition between the internal material (silica core) and the conic surface (Al thin film) of the probe. Moreover, a thinner coating can excite a larger number of surface plasmons which interact with the incident field and form the localized fields inside the probe tip. This is to be expected since many surface charge densities will be induced in this metal thin-film coating by the incident electric fields.

3.4 Comparison of 3D NSOM images of dielectric tip and metal tip

On the basis of our simulations for the metal-coated tip, the electric field is found to decay rapidly along the transverse and longitudinal directions as compared to the noncoated tip (the results are not shown here). The depolarization phenomenon of electric field components is the near-field effect. The characteristics of edge enhancement of field intensity deteriorate as the distance from the tip apex increases. For the metal-coated tip, this phenomenon exists several ten nanometers away from the tip aperture. A comparison of the field distributions decaying along the z -axis reveals that the field distribution of the metal-coated tip decays faster than that of the noncoated tip. For the metal-coated tip, the range of the field distribution decaying

along the z -axis is about 40 nm at $1/e$ of the maximum value, and for the noncoated tip, it is about 300 nm. This is because of the different waveguide structures and the different boundary conditions of the tip apices. In the case of the noncoated tip, the HE_{11} can propagate in any sizes of cone-shape tip, since there is no clear cutoff plane to separate from the radiation wave and evanescent field. As regards to the coated tip, the propagation mode has changed because of the existence of the metal film and formed the near-field coupling into the forbidden zone. Due to the shielding effect of the metal film, the presence of a metal coating on the tip prevents the lateral escape of light and concentrates most of it inside the tip. This light is well localized and originates from evanescent components exclusively. Moreover, the metal-coated tip possesses the phenomena of optical enhancement and beam confinement due to the effects of the surface atoms in the region of nanodimensions, i.e., the dipole polarization on the surface of the aperture rim and the beam blocking by the metal film of the conic surface.

Therefore, knowledge of the polarization is very important for the interpretation of images. The noncoated tip produces a well-confined and intense central spot from s-polarization, with a central decay due to the depolarization effects occurring in p-polarization. For the metal-coated tip, p-polarization is responsible for surface digging. As shown in Figs. 6(a) and 6(b), the intensity of p-polarization is the most important one. In Figs. 6(c) and 6(d), we see that less light is generated by the tip apex in s-polarization. In contrast, in p-polarization, we see more light at aperture rim, coming from the metal-coated tip and forming a local dipole along the aperture rim.

3.5 New design for fabricating an optimal tip

In this section, we present a new technique that combines a metal-coated probe with a dielectric tip. Here, we can adopt both the advantages of high transmission efficiency with no absorption of the dielectric tip and local enhancement of the metallic coating (the tip coated with a metallic thin film which could be used to produce spots smaller than those generated with the dielectric tip) to design a new type of tip. We illustrate two types of improved tip, types A and B, in this section. Type A is a metal tip that combines with a local dielectric tip, as shown in Fig. 7(a). In the process of coating the metal tip with an aluminum thin film (by the evaporation process), we keep the tip apex exposed to the fabricated local noncoated dielectric tip. That is, the tip

Fig. 7. Sectional plane diagram of 3D improved tips: (a) the metal-coated tip combines with the dielectric tip and (b) the same as (a), except that a hollow tip apex forms a semispherical lens surface.

Fig. 8. Distribution of the 3D electric field intensity modulus around the tip-sample coupling zone (improved tip) for p-polarization: (a) in x - z plane (at $y = 70\Delta$), (b) in y - z plane (at $x = 70\Delta$), (c) in x - z plane (at $y = 70\Delta$), and (d) in y - z plane (at $x = 70\Delta$). The electric field originates from an incident wave coming from the left side.

bases on the metal-coated tip and extends the internal dielectric material outside the metal-coated tip. The diameter of the tip apex is 50 nm (10 cells) and the length of the noncoated part is 25 nm (5 cells). In front of the probe, at a distance of 15 nm (3 cells), a semi-infinite silicon medium is taken as the sample with a refractive index of $n = 1.5$. The dimensions of each cell are $\Delta x = \Delta y = \Delta z = \Delta = 5$ nm, and the entire region model consists of $140(x) \times 140(y) \times 140(z)$ cells. Type B is the same as type A, except that in type B, a hollow tip apex with a radius of 25 nm forms a semispherical lens surface as shown in Fig. 7(b). Figures 8(a)–8(d) show the calculated results in the x - z and y - z sectional planes by p-polarization illumination; in particular, Figs. 8(a) and 8(b) correspond to type A, while Figs. 8(c) and 8(d) correspond to type B. It can be seen in Figs. 8(a) and 8(b) for type A that the light emerges from the top of the metal-coated part into the dielectric medium of tip apex end. It is expected that the optical transmission will be enhanced at the interface between the coated and noncoated parts of the tip apex. The field-enhanced effect is due to the dipole polarization on the metal aperture rim and the beam blocking by the metal layer (coated part). The high transmission effect at the top of the tip apex is due to the effect of the nonabsorptive properties of the dielectric medium (non-coated part). Because of the use of both metal-coated and noncoated parts, the light transmission from the tip apex is higher than that of the noncoated tip as shown in Fig. 3(a) and the spot size is smaller than that of the metal-coated tip used alone as shown in Fig. 3(b). These results lead to the improvement of transmission and the decrease in the spot size of the light emerging from the tip apex. The spot size is

about $200 \times 75 \text{ nm}^2$ as shown in Fig. 8(a) in the x - z cross sectional plane for p-polarization illumination, which is smaller than that produced by the metal-coated tip (about $300 \times 100 \text{ nm}^2$, as shown in Fig. 6(a), in the x - z cross sectional plane for p-polarization illumination). Note that a higher transmission is observed. We observed that the field intensity of spot size shown in Fig. 8(b) is obviously higher than that shown in Fig. 6(b). The disadvantage of type A is a challenge for users to reach the accuracy requirements for fabricating a tip shown in Fig. 7(a).

As regards to type B, the light coming from the base of the objective lens (semispherical at the tip apex) is focused at output end of the tip apex in front of the sample surface. Because of the use of a semispherical lens at the tip apex shown in Fig. 7(b), the focus spot at the hollow tip apex is expected to be smaller than that of the metal-coated tip used alone as shown in Fig. 3(b), thus causing a smaller spot emerging from the tip apex. Due to the small distance (15 nm) between the tip and sample, a smaller spot is produced. The spot size is reduced to $70 \times 30 \text{ nm}^2$ as shown in Fig. 8(c) in the x - z cross sectional plane for the p-polarization illumination case, which is much smaller than those produced by type A and the metal-coated tip used alone. Using type B, we can obtain a very small spot size of $70 \times 30 \text{ nm}^2$, which allows us to realize a recording density of approximately 300 Gb/in^2 . Moreover, it is possible to reduce the recording marks below the laser beam spot by controlling the input beam power. The disadvantage of type B is also a challenge similar to the case of type A, particularly for the fabrication of a hollow tip apex shown in Fig. 7(b). A comparison of the intensities of field distribu-

Fig. 9. Comparison of field intensities along the y-axis direction, where the dotted line represents type A, solid line represents type B, and dashed line represents the metal-coated tip used alone.

tion between the two improved tips (types A and B) and metal-coated tip used alone was performed. Figure 9 shows the intensity of field distribution along the y-axis direction, where the dotted line represents type A, solid line represents type B, and dashed line represents the metal-coated tip used alone. It can be observed in Fig. 9 that type A exhibits a stronger field intensity than the other two types of tip. That is, the smaller beam spot size obtained using type B is achieved at the expense of field intensity.

On the basis of these simulation results, we can conclude that this two types of improved probe are better structures for our applications since they exhibit the advantages of both high throughput light efficiency and small beam spot size, and yield a better type of tip pattern from the viewpoint of fabrication.

4. Conclusion

A near-field analysis was carried out to obtain more insight on the mechanisms responsible for an NSOM architecture using the 3D FDTD method. Among the many types of structures simulated in this study, including an aperture on an infinite metallic thin plate with and without sample interactions, comparisons of noncoated and coated tips, a novel probe structure combining a metallic probe with a local dielectric tip, and a structure using a semispherical lens at the tip apex were carried out. In our simulations, we determined some characteristics of 3D NSOM structures, allowing us to realize more clearly the physical mechanism in NSOM. Note that the depolarization phenomenon of electric field components is important in the near-field zone. The phenomena of optical enhancement and beam confinement are due to the effect of surface atoms in the region of nanodimensions, i.e., the dipole polarization on the surface of the aperture rim and the beam blocking by the metal film of the conic surface. We also verified the FDTD designs of field-enhancing NSOM probes that are experimentally feasible and provided a suggestion for fabricating an optimal probe. Using the improved tips with a size of 50 nm, we can obtain a very small spot size of approximately the same dimensions as the tip apex, which is much smaller than those

produced by the noncoated or metal-coated tip used alone. In the near future, the nonlinear response of the irradiated sample and the NSOM application using photonic crystals will be further studied.

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