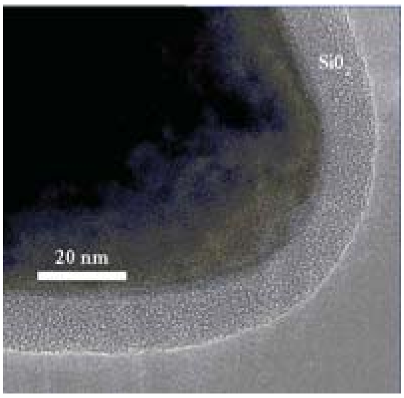
Scientific COMPUTING Information Technology for Science

Electromagnetic Simulation Helps to Optimize Optical Microscope Resolution

A new tip design delivers enhanced sub-20 nanometer performance

Erik J. Sanchez, Ph.D.

Near-field scanning optical microscopy (NSOM) has extended optical measurements past the diffraction limit, making it possible for the first time to view objects and features in the 50 to 100 nanometer (nm) range. Recent research has demonstrated that the use of apertureless probes can further improve spatial resolution to below 25 nm.[1-3] The next challenge is optimizing the tip design in order to strongly illuminate the sample at distances from the aperture that are hundreds of times closer than the dimension of the wavelength of the light that is employed. Trial and error methods are highly undesirable because of the great challenges involved in building and testing optics at nanometer scales. Researchers at Portland State University (PSU), led by Erik Sánchez and funded by the NSF[4] are overcoming this problem by using a commercial finite different time domain electromagnetic simulator (XFDTD) to analyze tip performance without the need to build a physical model.



click to enlarge

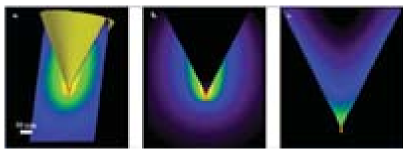
Figure 1: Transmission electron micrograph of a metal probe tip coated with 10 nm of electron beam-grown SiO_2 to prevent fluorescence quenching by the metal

Recent advances in nanotechnology and nanoscience are highly dependent on our newly acquired ability to measure and manipulate individual structures on the nanoscale. A drawback of light microscopy is the fundamental limit of the attainable spatial resolution dictated by the laws of diffraction at about 250 nm. This diffraction limit arises from the fact that it is impossible to focus light to a spot smaller than half its wavelength. The challenge of breaking this limit has led to the development of NSOM. The optical probes originally used in NSOM were created by pulling an optical fiber to a final diameter of 25 to 100 nm, coating it with aluminum, and etching to provide a flat, circular endpoint and aperture. Unfortunately, only a tiny fraction of the light coupled into the fiber is emitted by the aperture because of the cutoff of propagation by the waveguide modes. The low light throughput and finite skin depth of the metal limit the resolution to normally 50 to 100 nm.

Need for finer spatial resolution

Many applications require spatial resolutions that are not attainable with the aperture technique. For example, a spatial resolution of at least 30 nm is desirable in spectroscopic imaging of photosynthetic membranes in order to resolve closely packed individual proteins in a lipid membrane. This has been accomplished by the use of laser-illuminated metal tips to provide a local excitation source for the spectroscopic response of the sample under investigation. Excitation light of proper polarization induces a strongly enhanced field at the tip. The highly localized excitation source has provided resolution levels of 15 to 25 nm, making it possible, for example, to resolve tightly packed chromophoric membrane proteins.

Unfortunately, the presence of the metal tip nanometers away from the fluorophore leads to fluorescent quenching. This results is a negative fluorescent image, essentially a dark spot at the chromophore position surrounded by a sharp halo of emission. Sánchez, along with colleagues at Harvard University, had overcome this problem by depositing a homogenous, nanometer-scale silicon oxide coating on near-field probes in order to minimize quenching.[5] An important advancement was achieved when electron beam assisted deposition (EBAD) demonstrated the ability to evenly deposit silicon oxide coatings on the complex three-dimensional tips of the apertureless metal probes, which are typically only a few nanometers wide, in order to avoid fluorescent quenching. They used a focused ion beam (FIB)/scanning electron microscope (SEM) to grow dielectric material on a complex three-dimensional optical probe. A DualBeam FIB/SEM manufactured by FEI (Strata DB-235) uses a liquid metal gallium ion source accelerated to 30 kV with specimen currents from 1 pA to 20 nA. The researchers wrestled with and finally overcame the challenge of maintaining the integrity of the coating while achieving reasonably fast deposition rates. Using electron beam-aided deposition they were able to create a dielectric film with nanometer conformity over complex optical geometries.



click to enlarge

Figure 2: FDTD modeling of a sharp TENOM probe tip, (a) the 3-D view of the probe tip with crossection of E the external electric field magnitude, (b) the 2-D side view of the E field distribution, (c) and J, the current density magnitude.

Challenges of tip design

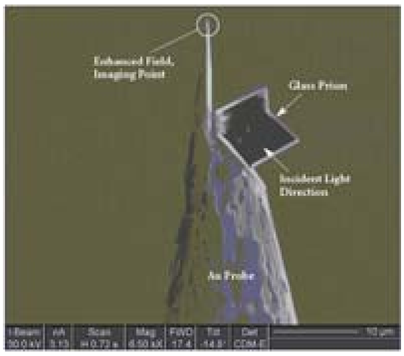
When a horizontally incident (vertical polarization) is applied, this probe experiences an increase of local field intensity (|**E**|) of ~1000 times.

The next step was optimizing the metal tip design. This was accomplished by modeling Chance Prock Silbey (CPS) theory for an emitting dipole aligned in close proximity to a metal substrate and then applying to a metal tip.[6] In detecting the fluorescent signal from a molecule, the optimum distance from the chromophore to the metal tip is approximately 22 nm for a parallel emitting dipole, or 24 nm for a perpendicular emitting dipole.[7] If the metal tip is closer than 3 to 5 nm, even with the coating, then the fluorescent signal is dramatically quenched, making it impossible to detect the signal. This sets up the primary challenge in tip design, optimizing the near-field tip design to deliver the highest possible electromagnetic field at the appropriate distance. Although the researchers believed the tip molecule separation would be similar to the bulk metal-molecule situation, the tip's geometry required careful modeling in order to verify this information. It would be very difficult and expensive to accomplish this objective using conventional build-and-test methods. The primary problem is the cost of fabricating the tips and the difficulty of performing field measurements at the nanometric scale.

For these reasons, the researchers used FDTD to model the near-field response of proposed designs. Electromagnetic simulation takes only a small fraction of the time and expense involved in building and testing apertureless tips. Simulation also provides more information than physical experiments by yielding results at every point in the solution domain, far exceeding the results that can be achieved with physical measurements. The researchers selected XFDTD software from Remcom, State College, PA, because it can quickly and reliably turn complicated geometries into accurate electromagnetic meshes. This ability is extended with the addition of an advanced meshing algorithm that makes meshing of certain difficult geometry features possible. Adaptive meshing capabilities reduce solution times while maintaining high levels of accuracy by automatically adjusting the mesh to provide more cells in areas with high transients and reducing cells in areas where there is less variation. In addition, the use of a parallel computational code allows for multiple computers to be connected in order to perform calculations faster as well as use larger workspaces.

Using simulation to iterate to an optimized design

The researchers began by simulating an existing tip design in order to validate the accuracy of the method. They divided the initial design into cubic cells with the appropriate frequency-dependent behavior. An absorbing boundary condition was used. The incident excitation field was set to an electric field strength of 1 V/m at 800 nm wavelength, and the space was discretized with 3 nm cubes. The material of the tip was defined to be Au and the spacer was SiO_2. The electromagnetic field in each cell under plane wave illumination was calculated by the software through time domain integration of Maxwell's equations. The intensity enhancement at the end of the tip is 100 without the spacer, approximately 5 nm in front of the tip. When the 9 nm SiO_2 spacer is present, the enhancement is reduced to 50. This is still high enough for near-field imaging at 5 nm in front of the tip. These results matched experimental measurements, so the researchers began using XFDTD in an effort to optimize the tip design.



click to enlarge

Figure 3: This novel NSOM probe takes advantage of a non-radiatively coupled focused spot of light, which creates a resonant plasmon that dramatically improves signal to noise. Light enters the prism, and the end of the tip creates a strong localized field.

Normally, apertureless near-field optical probes require direct illumination of the tip apex in order to generate a sub-diffraction limited light spot. A large background signal originates from the emission of many chromophores in the far-field illuminated volume. Typically, tips with a high field enhancement are used in order to overcome this background contribution. Another way to overcome this problem is to non-radiatively propagate a field to the end of the tip where the energy of the field could emit radiatively, eliminating the background contribution. Under certain conditions the energy carried by photons of light is transferred to packets of electrons, called plasmons, on a metal's surface. The light's energy is transferred to driving the electrons resonantly through attenuated total reflection at very specific conditions.

Guided by electromagnetic simulations, the researchers designed a tip that takes advantage of this technique, which was created by A. Otto in the late 1960s.[5] The angle of the prism, tip shaft length and gap between the prism and metal were carefully engineered to achieve resonance. When the plasmon reaches the tip end, it generates a strong evanescent field within a region on the order of the tip end diameter. Evanescent waves are formed when sinusoidal waves are internally reflected off an interface at an angle greater than the critical angle so that total internal reflection occurs. The intensity of evanescent waves decays exponentially as they move further from the interface at which they are formed. This eliminates the signal generated by far-field illumination, increasing the signal-to-background ratio. PSU researchers are currently evaluating the performance of these tips and working to improve their design to deliver even higher levels of performance.

References

1. Erik J. Sánchez, Lukas Novotny, and X. Sunney Xie, *Physical Review Letters* 82, 20, pg 4014-4017 May (1999).
2. Achim Hartschuh, Erik J. Sánchez, X. Sunney Xie and Lukas Novotny, *Physical Review Letters* 90, 9, 095503 March (2003).
3. Derek B. Nowak, John T. Krug II, X. Sunney Xie, and Erik J. Sanchez, *WMSCI Proceedings*, 9th World Multi-conference on Systemics, Cybernetics and Informatics, Orlando, Florida Conference, July (2005).

4. IDBR #0500812
5. E. J. Sánchez, J. T. Jrug, and X. S. Xie, *Review of Scientific Instruments* 73, 11, pg 3901-3906, Nov (2002).
6. John T. Krug II, Erik J. Sánchez and X. Sunney Xie, *Journal of Chemical Physics* 116, 24, June (2002).
7. John T. Krug II, Erik J. Sánchez and X. Sunney Xie, *Applied Physics Letters* 86, 233102, May (2005).

Erik J. Sanchez is an assistant professor of physics at Portland State University. He may be reached at editor@ScientificComputing.com.