Toward Complete Control of Localized Light and its Interactions with Matter

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Scientific Thrust Areas: (1) Plasmonic optical antennas and optical transformers, **(2)** Chemical imaging of plant biomass with μ-Raman spectroscopy for biofuel applications

Research Achievements

Plasmonic optical antennas and optical transformers - The recent invention of single metallic optical nanoantennas has greatly improved the mismatch between light and nanometer-scale objects[1]. Optical nanoantennas are specifically engineered to enhance fields at visible and near-infrared (NIR) wavelengths and confine them to nanoscale regions in size, significantly defeating conventional diffraction-limited resolution (~ 300 nm). By *efficiently* coupling light with localized plasmon modes, optical nanoantennas harness the short wavelength excitations to create extremely intense, ultra-confined optical fields.

One major thrust has been the fabrication and characterization of nanoantenna-based scanning probes that we will use for high-spatial resolution (nm-scale) imaging spectroscopy [fig. 1c]. Nanoantennas comprised of plasmonically-coupled metallic nanoparticles offer appreciable advantages over other near-field optical devices, most notably aperture-based and apertureless near-field scanning optical microscope probes. In particular, nanoantennas present much greater near-field coupling efficiency than NSOM probes (efficiency > 10% for a bowtie versus $\sim 10^{-5}$ for a typical metal-coated pulled fiber probe) and significantly higher field enhancement than has been measured for apertureless tips.

Another related research thrust has been the experimental and theoretical study of a device, termed the Plasmonic Color Nanosorter (PCoN), which demonstrates both the ability to efficiently capture and strongly confine broadband optical fields, as well as to spectrally filter and steer them while maintaining nanoscale field distributions [fig. 1a-b]. A central goal of plasmonics is complete control over optical signals at deeply sub-wavelength scales. The recent invention of optical nanoantennas has led to a number of device designs that provide confinement of optical fields at nanometer length scales. For photonic applications, however, the effectiveness of these structures would be *significantly improved* by the added ability to spatially sort the optical signals based on energy/color. Because of the capability to localize *and* steer photons, color nanosorters are expected to have profound impact on a wide range of optoelectronic and plasmonic applications including ultrafast color-sensitive photodetection, solar power light harvesting, and multiplexed chemical sensing.

In addition to the nanoantenna tips and the PCoN, we have undertaken a number of other plasmonics-related projects in our lab, including the study of novel tapered 3-dimensional nano-optical transformers/antennas specifically engineered for SERS detection (with users M. Wu and E. Yablonovitch, UCB), nonlinear optical imaging spectroscopy of plasmonic nanostructures [fig. 1d], and the demonstration of a MEMS-based tunable-gap nanoantenna fabricated with Au-coated TiOx nanoswords (with users B. Sosnowchik and L. Lin, UCB).

Chemical imaging of plant biomass with μ-Raman spectroscopy - A detailed knowledge of the plant cell wall will allow researchers to visualize the chemical and physical obstacles to biomass breakdown. This will be vital in developing better pretreatment procedures, and identifying plant material that may naturally be better suited to use in such processes. We demonstrate, for the first time, the label-free *in situ* study of wild type and low lignin transgenic poplar wood by confocal Raman microscopy aimed at the characterization and comparison of local compositional and structural traits. Accurate investigation of these samples will have significant impact on biofuel technology since low lignin wood offers potentials for improved biomass conversion into fermentable sugars. Importantly, lignin was found to be significantly reduced in the S2 wall layers of the transgenic poplar fibers, suggesting that such transgenic approach may help overcome cell wall recalcitrance to wood saccharification [fig. 1e-f]. Also, spatial heterogeneities in the lignin composition, particularly with regard to coniferaldehyde moieties, were found in both the wild type and the transgenic samples.

Future work: Our future goals include the use of nanoantenna scanning probes for nanoscale optical imaging spectroscopy of novel nanomaterial-based light-harvesting devices, non-invasive imaging of fundamental nanoscale mode volumes/field distributions created by plasmonic devices/structures, nano-Raman chemical mapping of plant cell walls, and the use of a novel background fluorescence-suppressing method for the first ever Raman chemical imaging studies of *Arabidopsis*.

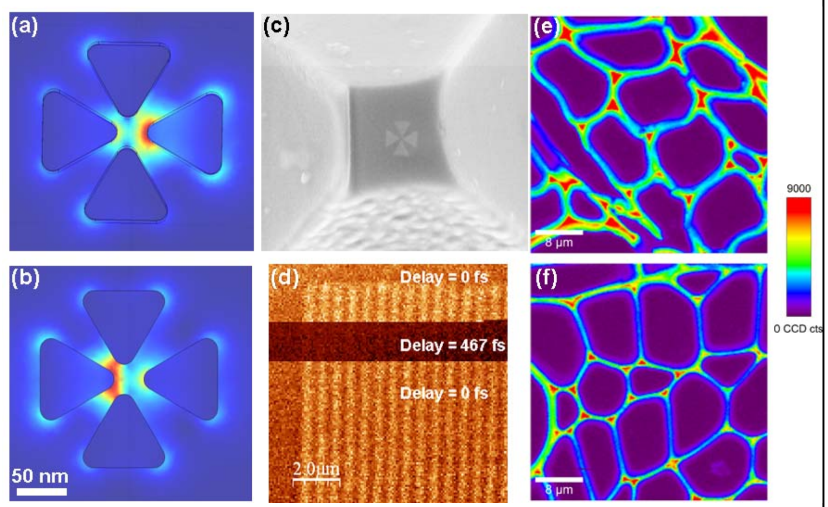

Figure 1: PCoN optical field distribution for (a) 780 nm excitation and (b) 820 nm excitation; (c) SEM image of a double-bowtie "cross" nanoantenna fabricated at the end of an AFM tip; (d) image of enhanced four-wave-mixing signal originating from a Au plasmonic grating. Note the disappearance of signal when pump and probe pulses are separated by a non-zero delay; chemical map of lignin distribution in wood from (e) wild-type poplar and (f) transgenic low-lignin poplar.

References:

[1] P. J. Schuck**,** *et al*., "Improving the mismatch between light and nanoscale objects with gold bowtie nanoantennas." *Phys. Rev. Lett.* 94, 017402 (2005).

Publications:

- S. Wu[†], Gang Han[†], *et al*, "Non-blinking and photostable upconverted luminescence from single lanthanide-doped nanocrystals," *PNAS*, in press (2009)
- M. Schmidt, *et al*., "Label-free *in situ* imaging of lignification in the cell wall of low lignin transgenic *Populus trichocarpa*," *Planta Rapid Comm*., submitted (2009)
- B. D. Sosnowchik, *et al*, "Tunable Optical Enhancement from a MEMS-Integrated TiO2 Nanosword Plasmonic Antenna," *Proceedings of 22th IEEE Micro Electro Mechanical Systems Conference*, pp. 128-131, Sorrento, Italy, Jan. 2009.
- C. Nobile, *et al*, "Probe tips functionalized with nanocrystal tetrapods for high resolution atomic force microscopy imaging," *Small* 4, 2123 (2008)
- A. Jamshidi, *et al*, "Dynamic manipulation and separation of individual semiconducting and metallic nanowires," *Nature Photonics* 2, 85 (2008), **Cover Article**
- F. Wang, *et al*, "Observation of excitons in one-dimensional metals," *Phys. Rev. Lett.* 99, 227401 (2007)