Towards the holy grail of near field optics via reproducibly fabricated optical nano antennae on Scanning Probe Tips

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The concept of optical antennae to reproducibly focus light well below the diffraction limit, while enhancing the optical near field several orders of magnitude [1, 2] has only been feasible due to the advancements in nano fabrication over the past few years. A lot of work has been done on various nano fabrication approaches [3] of optical antennae and the determination of their resonance behavior[4, 5], leading to novel mode-coupling schemes[5], antenna-coupled plasmonic waveguides [6, 7], and resonators that separate near fields energetically as well as spatially on length scales well below the diffraction limit[8].

Currently, significant effort has gone into using optical antennae for high-resolution sensing applications and for investigating matter optically on the nm scale via antenna-based near field probes[3, 9-12]. The latter has to overcome additional fabrication challenges since a functioning antenna must be placed reproducibly on the apex of a scanning probe tip with a feature resolution on the order of 10nm and a placement precision on the order of 5nm.

The most promising optical antenna design for near-field imaging purposes are plasmonically-coupled structures. These types of antennae take advantage of the ultra-enhanced and localized fields created around the nanogaps or crevices between constituent substructures, and are responsible for the largest reported scattering and fluorescence signal enhancements to date (e.g. single-molecule surface-enhanced Raman scattering).

We employed Focused Ion Beam Milling and Induced Deposition Mask Lithography[3] to fabricate a variety of promising coupled optical antennae on scanning probe tips and successfully demonstrated their reproducible functionality: We showed record-breaking Raman signal enhancements on the order of ~1000 with bowtie-like antennae, measuring the tip-down versus tip-up signal ratio as the figure of merit, without the need for the rather limiting metal-substrate-mediated gap mode. Coaxial optical antenna tips enabled the recording of useful Raman spectra in ~50 ms to acquire 256 by 256 pixel images on dielectric substrates with a full spectrum at each pixel [4]. This technique, also referred to as hyperspectral imaging, has lead to full chemical maps of the investigated Carbon Nanotube bundles with a resolution below 20nm. More recently, we developed a novel geometry which are bell tower like “campanile” tips at the end of optical fibers that we will present here for the first time, enabling nano-photoluminescence hyperspectral imaging on Indium Phosphide nanowires. The important advancement of this structure is the **capability of exciting and collecting through the optical fiber with high efficiency**, precluding the need for a transparent substrate and is combining almost all features the near field optics community is wishing for in terms of an ideal near field probe.

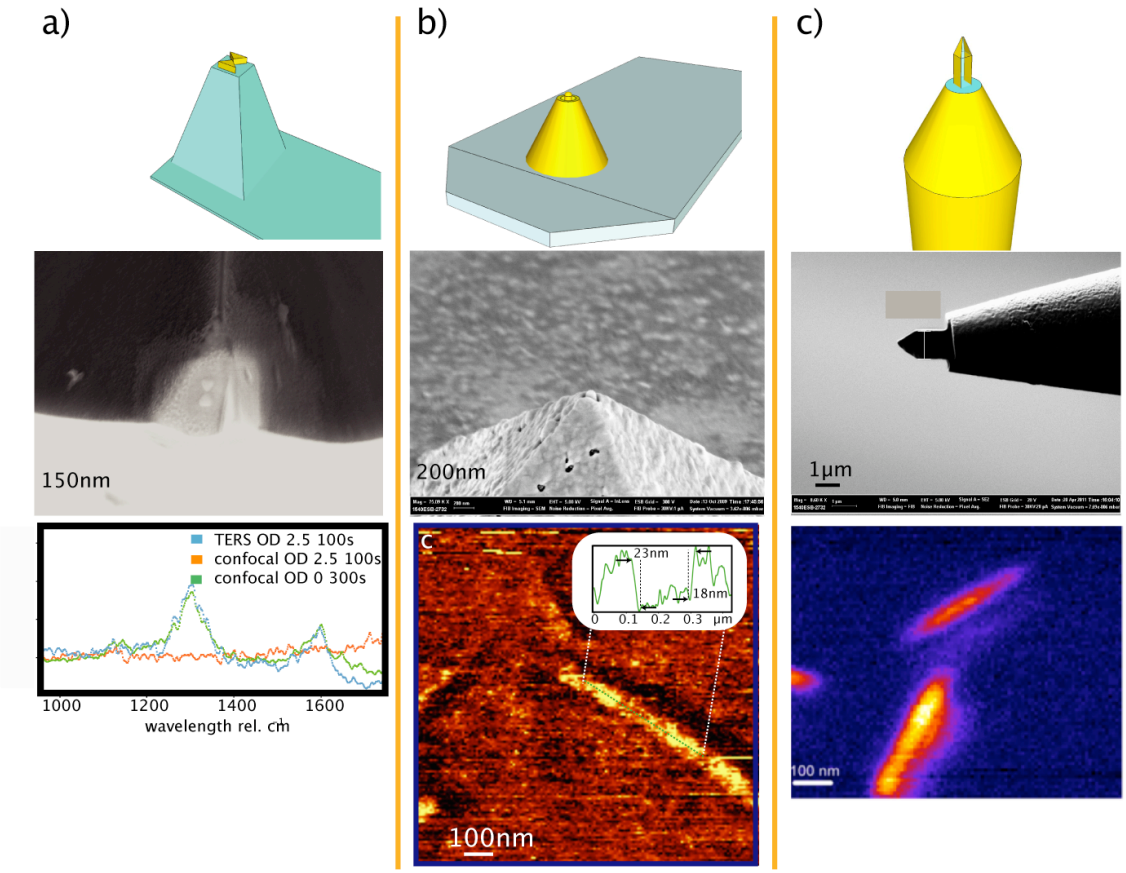

Figure 1 various coupled optical antennae structures fabricated on Scanning Probe tips. a) Au bowtie antennae on SiN tips leading to 800-fold signal enhancement of the CNT Raman signal via Tip Enhanced Raman Spectroscopy. b) coxial optical antennae on SiN tips enabling hyper spectral imaging showing the D-mode chemical map of CNT bundles. c) bell tower like "campanile" tip on a optical fiber, enabling nano photo luminescence on Indium Phosphide nano wires, where both excitation and collection is done through the optical fiber.

1. Schuck, P. J.; Fromm, D. P.; Sundaramurthy, A.; Kino, G. S.; Moerner, W. E. *Physical Review Letters* **2005,** 94, (1), -.
2. Muhlschlegel, P.; Eisler, H. J.; Martin, O. J. F.; Hecht, B.; Pohl, D. W. *Science* **2005,** 308, (5728), 1607-1609.
3. Weber-Bargioni, A.; Schwartzberg, A.; Schmidt, M.; Harteneck, B.; Ogletree, D. F.; Schuck, P. J.; Cabrini, S. *Nanotechnology* **2010,** 21, (6), -.
4. Weber-Bargioni, A.; Cornaglia, M.; Dhuey, S.; Ogletree, D. F.; P.J., S.; Cabrini, S. *submitted*
5. Zhang, Z.; Weber-Bargioni, A.; Wu, S. W.; Dhuey, S.; Cabrini, S.; Schuck, P. J. *NANO Letters* **2009,** 9, (12), 4505-4509.
6. de Waele, R.; Burgos, S. P.; Atwater, H. A.; Polman, A. *Opt. Express* **2010,** 18, (12), 12770-12778.
7. de Waele, R.; Burgos, S. P.; Polman, A.; Atwater, H. A. *Nano letters* **2009,** 9, (8), 2832-2837.
8. McLeod, A.; Weber-Bargioni, A.; Zhang, Z.; Dhuey, S.; Harteneck, B.; Neaton, J. B.; Cabrini, S.; Schuck, P. J. *Physical Review Letters* **2011,** 106, (3), 037402.
9. Farahani, J. N.; Pohl, D. W.; Eisler, H. J.; Hecht, B. *Physical Review Letters* **2005,** 95, (1), 017402.
10. Weber-Bargioni, A.; Schwartzberg, A.; Cornaglia, M.; Ismach, A.; Urban, J. J.; Pang, Y.; Gordon, R.; Bokor, J.; Salmeron, M. B.; Ogletree, D. F.; Ashby, P.; Cabrini, S.; Schuck, P. *Nano letters* **2011,** 11, (3), 1201-1207.
11. De Angelis, F.; Das, G.; Candeloro, P.; Patrini, M.; Galli, M.; Bek, A.; Lazzarino, M.; Maksymov, I.; Liberale, C.; Andreani, L. C.; Di Fabrizio, E. *Nat Nanotechnol* **2010,** 5, (1), 67-72.
12. Hoppener, C.; Novotny, L. *Nanotechnology* **2008,** 19, (38), -.