Supplement to "**Toward Nanometer-scale Optical Photolithography: Utilizing the Near-Field of Bowtie Optical Nanoantennas**"

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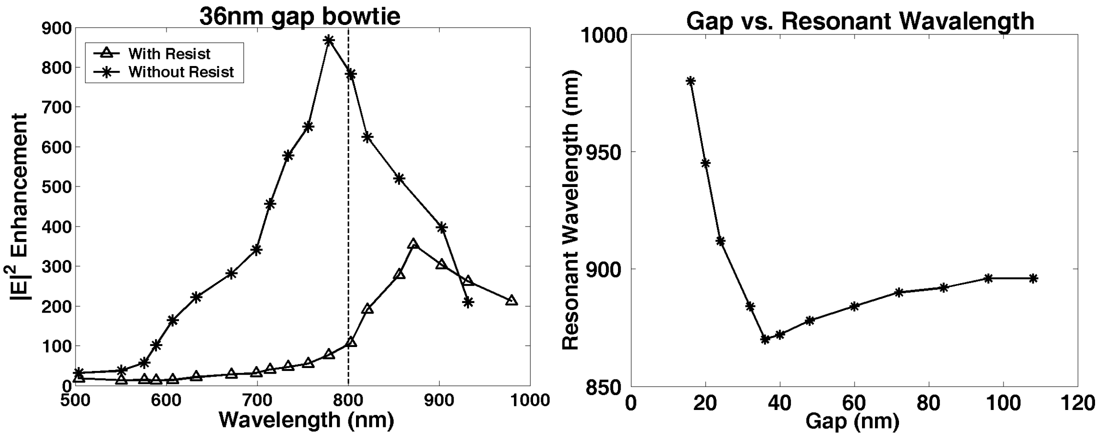
## Finite Difference Time Domain(FDTD) Simulations

Along with the experimental measurements, we also studied the field enhancement and confinement in bowtie antennas by FDTD simulations, as previously reported[2,4]. The FDTD simulations for this report modeled bowtie antennas that closely resemble the fabricated structures and were covered with a 76nm layer of SU-8 resist, whose optical properties are known at the pump wavelength (800nm). From the SEM images, we observed that our best resist exposures (at 27 μW and 54 μW) were from bowties with larger gap widths (~36nm). The results from the FDTD computations for a 36nm gap width bowtie are shown in Fig 3. The FDTD simulations predict an intensity enhancement of 107, at an excitation wavelength $\lambda$ = 800 nm, 4nm above the surface of the antenna. This field enhancement is confined to spots close to the tip of either triangle in the bowtie and the intensity enhancement falls off to ~16 in the gap between the triangles. The profile of the resists features for the 27 μW case, a direct measure of confined field enhancement and distribution, correlates well with rigorous FDTD simulations of field distribution.

We also characterized the enhancement from bowtie antennas by simultaneously measuring TPPL from them during the exposure. We described the process of obtaining the intensity enhancement from TPPL measurements in our earlier report[3]. The enhancement figure obtained from TPPL measurements for ~36 nm gap bowties were higher than those obtained from bowties with smaller gaps on the order of ~16nm. In our earlier work[2,4], we had analyzed the resonant behavior of bowtie antennas with different gap widths and concluded that the capacity of the air gap plays a dominant role in determining the resonant wavelength($\lambda_{res}$) for small gap widths. We observed that $\lambda_{res}$ first blue shifts with increasing gap and then red-shifts beyond a certain gap width[4]. The smaller gap bowties resonate at longer wavelengths (for a 16nm gap, $\lambda_{res}$=830nm). When the bowties are immersed in a dielectric like optical resist (refractive index, n=1.59) the resonant wavelength is significantly red-shifted compared to $\lambda_{res}$ in air. We thus observed from FDTD simulations that a 16nm gap bowtie covered with optical resist was a long way off the resonant wavelength ($\lambda_{res}$= 980nm) at $\lambda_{incident}$= 800nm compared to 36nm gap width bowtie($\lambda_{res}$=870nm, Intensity enhancement ~ 350). The spectra obtained from FDTD simulations for a 36nm gap bowtie covered with resist and without resist is given in Fig 4(a). The variation in resonant wavelength with gap width for a bowtie covered with resist is shown in Fig 4(b). Because we are constrained to work at wavelengths ≤ 800 nm to ensure sufficient TPA in the resist, the highest enhancements are observed for bowties with gaps ~35-40 nm rather than at the smallest gap sizes. This is expected and agrees well with FDTD modeling of the bowtie/resist system, which predicts that bowties with ~40 nm gaps will have the shortest wavelength resonances (in contrast to the bowtie-in-air case, where the shortest wavelength resonance with Au bowties occurs at a gap size of ~60 nm). Hence we see our best resist exposures and highest TPPL enhancements for ~36nm gap width bowties. Thus the resonant antenna effect in bowties is clearly evident in the experimental measurements and FDTD simulations. In the future, optical antennas designed with "bluer" resonances corresponding to efficient TPP wavelengths (using, e.g., smaller constituent triangles or different antenna material such as Ag) will help optimize nanoscale lithography.



(a) (b)

Fig 4: (a) FDTD simulations of a 36nm gap width bowtie covered with resist and without resist. The dotted line denotes the incident pump wavelength. The resonant wavelength ($\lambda_{res}$) is 872nm for a 36nm gap bowtie covered with resist. (b) Variation of resonant wavelength with gap width for a bowtie covered with resist.