Ultra-Narrow Gap Al/Al2O3 Bowtie Nanoantennas by Focused He Ion Milling Lithography

Massimo Cuscunà[a], Giuseppe Calafiore[b], Daniela Simeone[a], and Stefano Cabrini[b]

[a] *CNR-NANOTEC Institute of Nanotechnology, Lecce 73100, Italy*
[b] *The Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley CA, 94720, USA*
email: scabrini@lbl.gov

Plasmonic nanoantennas are interesting nanostructures, capable to manipulate the light in sub wavelength dimensions. Particularly bowtie antennas are able to achieve very high local electromagnetic (EM) field confinement and enhancement in the gap between the two triangles (hot spot). This effect is strongly dependent on the gap size, which in turn is related to technological limitations. Although several studies have been done on the effect of the gap between plasmonic antennas [1], here we present a comprehensive study on the structural and optical properties of aluminum bowties, realized with ultranarrow gap by He+-ion milling lithography, and operating from the near-infrared to the red part of the visible range [2]. In this study we discuss the effects of the fabrication process and the formation of the native aluminum oxide as a fundamental contribution for the optimization of the plasmonic antennas (figure 1). This analysis demonstrates that large EM near-field enhancement and different hot spot spatial positions, as a function of nanometer-sized gaps, are constrained by the native aluminum oxide, thus, working as hot spot ruler.

The combination of EBL and He+-ion milling provided a few nanometer milling precision; that allows the fabrication of extremely narrow gap Al bowtie nanoantennas generating EM hot spots at visible frequencies. Nanoscale structural investigation based on SEM techniques and optical characterization demonstrate that a 3 nm gap is the intrinsic lower limit of the proposed nanofabrication approach because of the Al2O3 native oxide formation (figure 2). We used Finite element method calculations to determine that large EM near-field enhancement and different hot spot spatial positions, as a function of nanometer-sized gaps, are strongly constrained by the native aluminum oxide (figure 3). Because of that, we can assert that a gap between freshly Al triangular nanoprisms slightly larger than 3 nm would provide the maximization of the EM field enhancement factor. Furthermore, FEM analysis and SNOM measurements demonstrated the presence of enhanced near-field in the red side of the visible spectral range by proper downscaling the bowtie nanoprism size. These results, combining the He+-ion nanofabrication capabilities, in terms of high resolution, accuracy and high nanostructure quality, with the choice of Al as plasmonic metal can be employed to develop advanced plasmonic biosensors, novel miniaturized nanophotonic systems as well as platforms to investigate strong light−matter interactions at visible frequencies. Strong and concentrated EM field produced by ultranarrow gap bowtie nanoantennas, operating at visible frequencies, can be of great impact in biosensing applications and in the study of strong light−matter interactions with organic molecules.

The work at the Molecular Foundry was supported by the Office of Science, Office of Basic Energy Sciences, of the U.S. DOE under Contract No. DE-AC02-05CH11231. We would like to acknowledge Frances I. Allen and Paul Lum at Biomolecular Nanotechnology Center/QB3, University of California, Berkeley, CA, for the use of the Carl Zeiss Orion Nanofab

[1] B. Song et al.: “Probing Gap Plasmons Down to Subnanometer Scales Using Collapsible Nanofingers”; ACS Nano, June 2017

[2] D. Simeone et al: “Tailoring Electromagnetic Hot Spots toward Visible Frequencies in Ultra-Narrow Gap Al/Al2O3 Bowtie Nanoantennas”; ACS Photonics, July 22, 2018

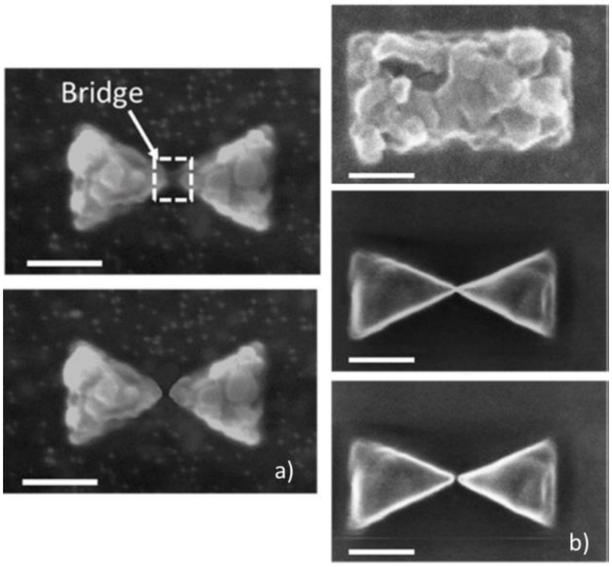

Figure 1. Helium ion microscopy images of Al bowtie nanoantennas fabricated by:
(a) two connected equilateral triangular nanoprisms produced by EBL technique (top) subsequently separated with HIL cut (bottom)
(b) EBL definition of a Aluminum rectangle (top); two-stage He+-ion milling, one for the triangles (center) and one for the gap (bottom). Scale bars are 100 nm.

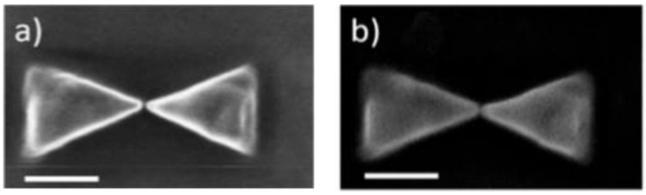

Figure 2. (a) HIM Image of a freshly fabricated bowtie dimer with geometrical parameters, triangle base and hight = 140 nm. (b) SEM image of the same bowtie dimer after being exposed 40 days to indoor air at ambient conditions.

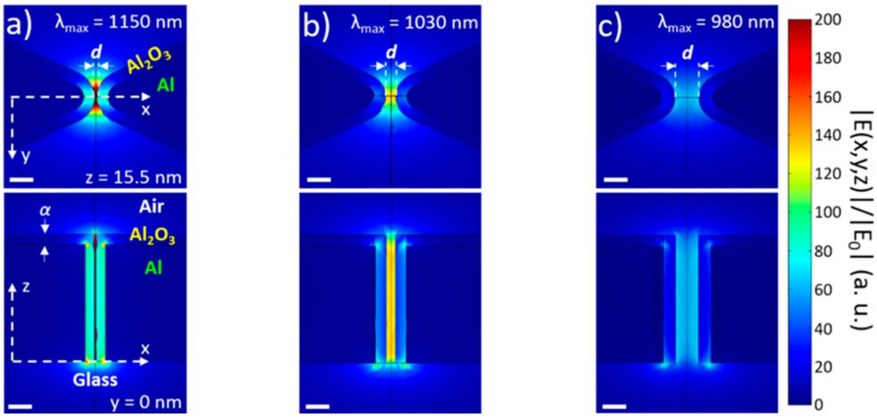

Figure 3. FEM simulation for bowtie dimers with different gap sizes: (a) 0.5, (b) 2, and (c) 5 nm: Calculated two-dimensional near-field enhancement factor |E|/|E0| (E0 = 1 V/m) distributions, at corresponding maximum wavelengths, in the Al/Al2O3 core−shell bowtie gaps (x−y) view (top panels) and (x−z) view (bottom panels)