## Plasmonic coupling across narrow gaps

F. Laible,<sup>1</sup> S. Dickreuter,<sup>1</sup> E. Gürdal,<sup>1</sup> F. Dai,<sup>1</sup> L. Seitl,<sup>1</sup> O. Hauler,<sup>2</sup> K. Braun,<sup>2</sup> A. Horneber,<sup>1,2</sup> D. Zhang,<sup>2</sup> P.-M. Adam,<sup>3</sup> A. J. Meixner,<sup>2</sup> D. Kern,<sup>1</sup> <u>M. Fleischer<sup>1</sup></u> <sup>1</sup>Institute for Applied Physics, <sup>2</sup>Institute of Physical and Theoretical Chemistry & Center LISA<sup>+</sup>, Eberhard Karls University Tübingen, 72076 Tübingen, Germany <sup>3</sup>L2n, Université de Technologie de Troyes, 10004 Troyes, France monika.fleischer@uni-tuebingen.de

Due to the continuous progress in nanofabrication techniques, coupling between two or more metallic nanostructures across narrow gaps can be ever better controlled. Under illumination, localized surface plasmon polaritons, in short plasmons, are excited in the nanostructures, which consequently act as antennas for visible light. By coupling neighboring structures across a gap of few ten nanometers down to Angstrom distances, several interesting effects occur.<sup>1</sup> High electric near-fields are strongly localized within the gap, making the system attractive e.g. for sensing or surface-enhanced Raman spectroscopy.<sup>2</sup> The coupling of the near-fields leads to mode hybridization. Energy splitting between bonding and antibonding modes can be observed. The spectral shift sensitively depends on the gap dimensions, therefore such systems can be employed as socalled plasmon rulers.<sup>3,4</sup> Few-nanometer gaps between contacted antennas can be bridged by molecules, allowing for combined electronic transport and optical investigations. If the gap is further reduced to sub-nanometer dimensions, tunneling effects occur,<sup>5</sup> giving access to the field of quantum plasmonics. In this presentation different approaches for creating coupled plasmonic nanogap antennas based on electron beam and helium ion beam lithography,<sup>2-6</sup> rapid thermal annealing,<sup>4</sup> vertical metal-insulator-metal structures,<sup>2</sup> or self-assembly<sup>7</sup> will be shown (cf. Figure 1). When the structures are prepared on flexible substrates an external strain can be applied, adding the possibility to reversibly tune the gap width and therefore the coupling strength (cf. Figure 2). $^{3,8}$ Simulations and measurements of the ensuing plasmon resonances, linear and non-linear spectral properties, and enhancing capabilities will be discussed.

<sup>&</sup>lt;sup>1</sup> F. Laible, A. Horneber, M. Fleischer, Advanced Optical Materials, submitted review (2021)

<sup>&</sup>lt;sup>2</sup> F. Dai, A. Horrer, P.-M. Adam, M. Fleischer, Advanced Optical Materials 8, 1901734 (2020)

<sup>&</sup>lt;sup>3</sup> F. Laible, D.A. Gollmer, S. Dickreuter, D. P. Kern, M. Fleischer, Nanoscale 10, 14915 (2018)

<sup>&</sup>lt;sup>4</sup> S. Dickreuter, D. P. Kern, M. Fleischer, Nanophotonics 7(7), 1317 (2018)

<sup>&</sup>lt;sup>5</sup> F. Laible, K. Braun, O. Hauler, M. Eberle, D. P. Kern, A. J. Meixner, M. Fleischer, Nanophotonics 9(6), 1391 (2020)

<sup>&</sup>lt;sup>6</sup> L. Seitl, F. Laible, S. Dickreuter, D. A. Gollmer, D. P. Kern, M. Fleischer, Nanotechnology 31, 075301 (2019)

<sup>&</sup>lt;sup>7</sup> E. Gürdal, A. Horneber, N. Shaqqura, A. J. Meixner, D. P. Kern, D. Zhang, M. Fleischer, Journal of Chemical Physics 152, 104711 (2020)

<sup>&</sup>lt;sup>8</sup> O. Hauler, L. Jakob, K. Braun, F. Laible, M. Fleischer, A. J. Meixner, F. Wackenhut, The Journal of Physical Chemistry C, accepted (2021)



*Figure 1: Examples of plasmonic nanogap antennas:* A) Gold nano-pillar dimer transformed into gold nanosphere dimer by rapid thermal anneal (RTA);B) bow tie nanoantenna; C) vertical gold-oxide-gold dimers;D) contacted bow tie antenna at the center of a break junction



*Figure 2: Dark-field spectra of a single bow tie antenna:* the bow tie is oriented perpendicular to the direction of strain on an elastomeric substrate. Compared to the spectrum without strain (black curve), when 25% strain is applied (red curve), necking of the substrate occurs, the gap size is reduced, coupling increases, and the dominant bonding mode is shifted to longer wavelengths