Optically active substrates through DNA-based lithography

Veikko Linko[1,2*]

[1]*Institute of Technology, University of Tartu, Estonia*
[2]*Aalto University School of Chemical Engineering, Finland*
**veikko.pentti.linko@ut.ee*

Self-assembling biotemplates such as the versatile and rapidly folding DNA origami[1,2] provide an effective shortcut for fabricating nanoscale assemblies with intricate, sub-nanometer detail. Despite their convenience, then converting these biological constructs into equally accurate and small structures with more favorable physical properties is significantly more challenging. One recent approach attempting to overcome this is DNA-assisted lithography (DALI).[3] DALI integrates DNA origami nanostructures as templates in negative-pattern lithographic masks and processes these with standard nanofabrication techniques (plasma etching, material evaporation, lift-off) to create metallic replicas of the template. Thus far, DALI has been used to pattern particles with sub-10 nm feature sizes from a limited selection of materials on silicon nitride and sapphire substrates.[3]

Here, we present a remarkably more adaptable method that circumvents the material constraints of DALI with the incorporation of a sacrificial polymer layer into the process flow. The new technique, biotemplated lithography of inorganic nanostructures (BLIN),[4] enables the pattern transfer of engineered biotemplates into inorganic structures from a multitude of materials and on less restricted substrates. Due to its versatile and parallel nature, BLIN could provide an attractive alternative for patterning optically active devices and other nanopatterned surfaces. To demonstrate this, we fabricate Ag bowtie nanoantennas on common glass substrates and characterize their response in the near-infrared regime using optical transmission measurements and FDTD simulations. Further, we use the patterned substrates in surface-enhanced Raman spectroscopy (SERS) to augment the detection of rhodamine 6G.[5]

Moreover, we can create even more efficient SERS supporting substrates, which are based on DALI and a layered configuration of materials. In detail, we use DNA origami templates to form hybrid nanostructures consisting of aligned silver bowtie-shaped particles and apertures of similar shape in a silver film. It was shown that this particular geometry facilitated a four-fold advantage in Raman enhancement compared to common particle-based SERS substrates (experiments and FDTD). Our DALI-fabricated hybrid structures suppress the background emission, allow emission predominantly from the areas of high field enhancement, and support additional resonances associated with the nanoscopic apertures. Finally, these nanoapertures also enhance the fields associated with the resonances of the underlying bowtie particles.[6,7] The versatility and parallel nature of our fabrication scheme and all above-mentioned features make our optically resonant substrates attractive for various SERS-based applications.

References

1. P. W. K. Rothemund, *Nature*, 2006, **440**, 297–302.
2. H. Ijäs, T. Liedl, V. Linko and G. Posnjak, *Biophys. J.*, 2022, **121**, 4800–4809.
3. B. Shen, V. Linko, K. Tapio, S. Pikker, T. Lemma, A. Gopinath, K. V. Gothelf, M. A. Kostiainen and J. J. Toppari, *Sci. Adv.*, 2018, **4**, eaap8978.
4. P. Piskunen, B. Shen, A. Keller, J. J. Toppari, M. A. Kostiainen and V. Linko, *ACS Appl. Nano Mater.*, 2021, **4**, 529–538.
5. K. M. Kabusure, P. Piskunen, J. Yang, M. Kataja, M. Chacha, S. Ojasalo, B. Shen, T. K. Hakala and V. Linko, *Nanoscale*, 2022, **14**, 9648–9654.
6. K. M. Kabusure, P. Piskunen, J. Yang, V. Linko and T. K. Hakala, *Nanoscale*, 2023, **15**, 8589–8596.
7. K. M. Kabusure, P. Piskunen, J. J. Saarinen, V. Linko and T. K. Hakala, *in preparation*, 2024.