P16 – DNA superstructures and origami shapes for the realization of metal nanoparticle pattern

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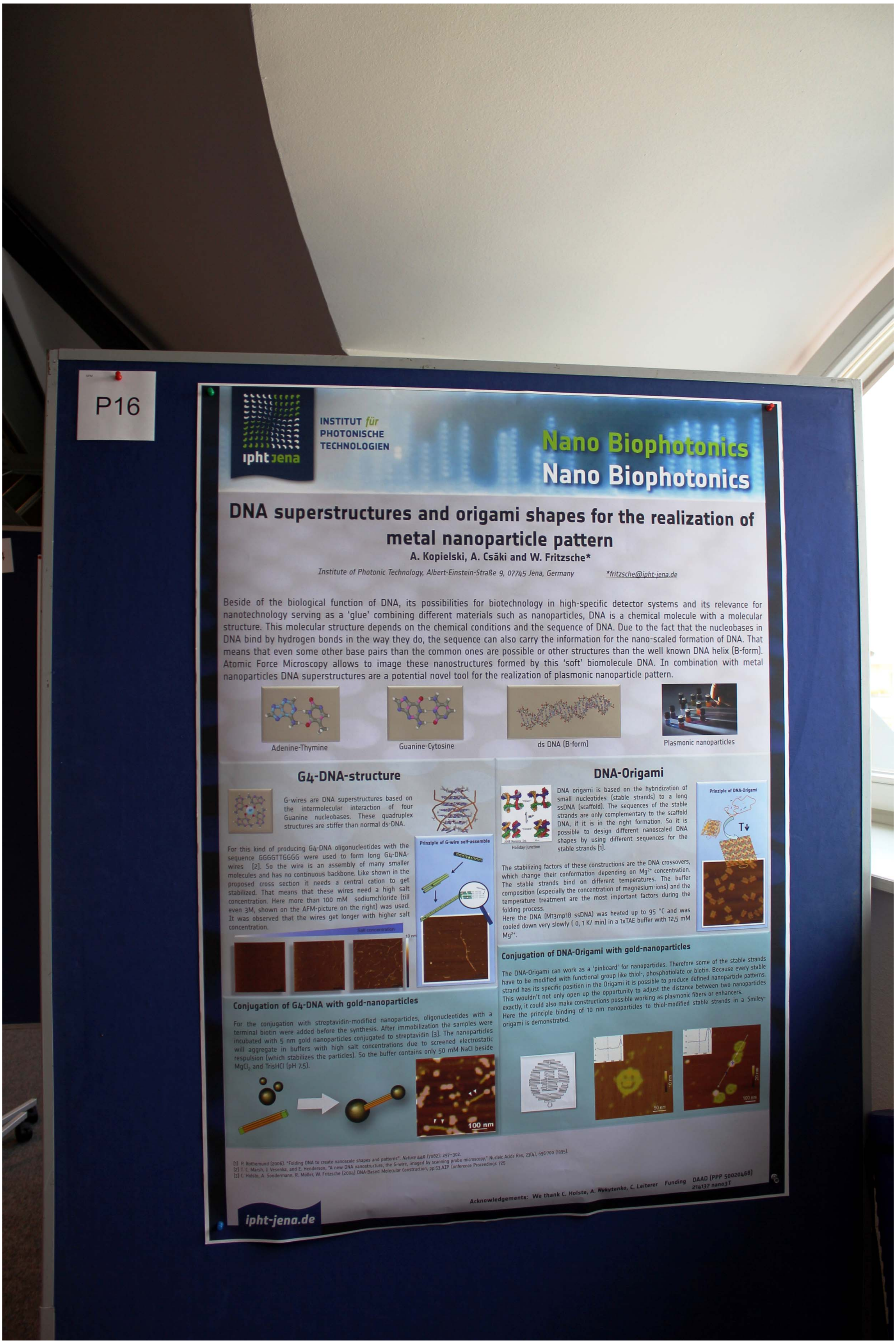
DNA is more than just the carrier for genetic information. Even in nature with its function as a matrix for specific binding biomolecules, some DNA-superstructures protect the end of the chromosomes and some small strands play an important role in catalytic or regulatory pathways. Because of their interaction properties and their size DNA molecules are especially interesting for molecular nanotechnology. The probably most noted method of DNA nanotechnology today is DNA origami [1]. Folding a long single strand of DNA aided by multiple smaller "staple" strands allows the creation of two- and three-dimensional shapes at the nanoscale. Guanine-quadruplex-structures, which protect the end of the chromosome from deterioration in nature, are another but not lesser interesting object for bionanotechnology. Guamine-rich oligonucleotides form longer four stranded G-Wires by self-assembly [2]. Metal-nanoparticle structures are interesting for their optical properties caused by plasmonic effects, with significant effects when particles get closer. Incident electromagnetic waves induce a collective oscillation of the conduction electrons in subwavelength particles. This excitation can couple with other particles in very short distances by plasmonic resonance effects. So in theory an accurately defined nanoparticle pattern could function as a “plasmonic fiber” or as an amplifier [3]. So the combination of DNA-superstructures with their features of nanoscaled definition options and metal nanoparticles is just the next logical step.

Here we present our experiments of the fabrication and specific binding of plasmonic nanoparticles to DNA-origami-shapes and G4-DNA-superstructures by using the self-assembly of DNA.

[1] P.Rothemund (2006). "Folding DNA to create nanoscale shapes and patterns". Nature **440** (7082): 297–302.

[2] T. C. Marsh, J. Vesenka, and E. Henderson, “A new DNA nanostructure, the G-wire, imaged by scanning probe microscopy,” Nucleic Acids Res, 23(4), 696-700 (1995).

[3] S.Bidault, F.J. Garcia de Abajo, A. Polman, Plasmon-Based Nanolenses Assembled on a Well-Defined DNA Template, J. Am. Chem. Soc.,2008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DNA superstructures and origami shapes for the realization of metal nanoparticle pattern
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Beside of the biological function of DNA, its possibilities for biotechnology in high-specific detector systems and its relevance for nanotechnology serving as a 'glue' combining different materials such as nanoparticles, DNA is a chemical molecule with a molecular structure. This molecular structure depends on the chemical conditions and the sequence of DNA. Due to the fact that the nucleobases in DNA bind by hydrogen bonds in the way they do, the sequence can also carry the information for the nano-scaled formation of DNA. That means that even some other base pairs than the common ones are possible or other structures than the well known DNA helix (B-form). Atomic Force Microscopy allows to image these nanostructures formed by this 'soft' biomolecule DNA. In combination with metal nanoparticles DNA superstructures are a potential novel tool for the realization of plasmonic nanoparticle pattern.
Adenine-Thymine
Guanine-Cytosine
ds DNA (B-form)
Plasmonic nanoparticles
G4-DNA-structure
G-wires are DNA superstructures based on the intermolecular interaction of four Guanine nucleobases. These quadruplex structures are stiffer than normal ds-DNA.
For this kind of producing G4-DNA oligonucleotides with the sequence GGGGTTGGGG were used to form long G4-DNA-wires [2]. So the wire is an assembly of many smaller molecules and has no continuous backbone. Like shown in the proposed cross section it needs a central cation to get stabilized. That means that these wires need a high salt concentration. Here more than 100 mM sodiumchloride (till even 3M, shown on the AFM-picture on the right) was used. It was observed that the wires get longer with higher salt concentration.
Prinziple of G-wire self-assemble
Salt concentration
10 nm
Conjugation of G4-DNA with gold-nanoparticles
For the conjugation with streptavidin-modified nanoparticles, oligonucleotides with a terminal biotin were added before the synthesis. After immobilization the samples were incubated with 5 nm gold nanoparticles conjugated to streptavidin [3]. The nanoparticles will aggregate in buffers with high salt concentrations due to screened electrostatic respulsion (which stabilizes the particles). So the buffer contains only 50 mM NaCl beside MgCl2 and TrisHCl (pH 7.5).
100 nm
DNA-Origami
DNA origami is based on the hybridization of small nucleotides (stable strands) to a long ssDNA (scaffold). The sequences of the stable strands are only complementary to the scaffold DNA, if it is in the right formation. So it is possible to design different nanoscaled DNA shapes by using different sequences for the stable strands [1].
Holiday-junction
Prinziple of DNA-Origami
T↓
The stabilizing factors of these constructions are the DNA crossovers, which change their conformation depending on Mg2+ concentration. The stable strands bind on different temperatures. The buffer composition (especially the concentration of magnesium-ions) and the temperature treatment are the most important factors during the folding process.
Here the DNA (M13mp18 ssDNA) was heated up to 95 °C and was cooled down very slowly (0, 1 K/ min) in a 1xTAE buffer with 12,5 mM Mg2+.
Conjugation of DNA-Origami with gold-nanoparticles
The DNA-Origami can work as a 'pinboard' for nanoparticles. Therefore some of the stable strands have to be modified with functional group like thiol-, phosphotiolate or biotin. Because every stable strand has its specific position in the Origami it is possible to produce defined nanoparticle patterns. This wouldn't not only open up the opportunity to adjust the distance between two nanoparticles exactly, it could also make constructions possible working as plasmonic fibers or enhancers.
Here the principle binding of 10 nm nanoparticles to thiol-modified stable strands in a Smiley-origami is demonstrated.
10 nm
50 nm
20 nm
100 nm
[1] P. Rothemund (2006). "Folding DNA to create nanoscale shapes and patterns". Nature 440 (7082): 297–302.
[2] T. C. Marsh, J. Vesenka, and E. Henderson, "A new DNA nanostructure, the G-wire, imaged by scanning probe microscopy," Nucleic Acids Res, 23(4), 696-700 (1995).
[3] C. Holste, A. Sondermann, R. Moller, W. Fritzsche (2004) DNA-Based Molecular Construction, pp.53,AIP Conference Proceedings 725
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