Optical control of hierarchical DNA-functionalized nanoparticle self-assembly on 2D surfaces

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Abstract. We investigated the effect of light on the DNA-driven self-assembly to form hierarchical patterns on two dimensional surfaces. We specifically focused on the self-assembly of DNA-functionalized quantum dots (QDs) onto DNA-functionalized glass substrate while illuminating the surface with a laser during coating process. The region illuminated with a green laser remained uncoated with red-emitting QDs while the non-illuminated region was successfully coated. Next, a red laser whose light cannot be absorbed by the red-emitting QDs did not avoid the DNA-driven self-assembly of the QDs onto glass substrate. Additionally, we demonstrated that silica nanoparticles that had been functionalized with DNA and are not able to absorb the light in the visible regime were again coated on the surface while being exposed to a green laser. These results prove that the absorption of the light is responsible for controlling the binding-unbinding process of QDs. Finally, we added DNA-functionalized green-emitting QDs onto the area that was not coated with red-emitting QDs under the green laser exposure. We observed successful coating of previously uncoated regions with the green QDs. This revealed the viability of our technique for building up hierarchical structures without using, sophisticated, or resource-intensive microfabrication methods.

1 Introduction

DNA-driven self-assembly is a reversible technique for controlled assembly of nanomaterials at the nanoparticle scale, utilizing the selectivity of DNA molecules [1]. An important characteristics of this method is its reversibility, which is achieved by regulating the temperature of the medium containing the nanoparticles. Since the complementary single-stranded DNA (ssDNA) forms hydrogen bonds with one another, nanoparticles possessing complementary ssDNAs on their surfaces bind each other. Lowering the temperature causes the nanoparticles to stick together, while raising the temperature causes them to separate [2,3].

Our recent research showed that light absorbed by gold nanoparticles can manage the binding and unbinding of DNAs linked to gold nanoparticles without externally increasing the temperature. We demonstrated that by applying light to the self-assembled nanoparticles, the surroundings of the nanoparticles heat up via optical excitation. We proved that structure of the self-assembled nanoparticle network can be tailored optically leading to a reversible control of the optical features [4].

Here, we extend the applicability of optical control of the DNA-driven nanoparticle self-assembly towards producing their patterned thin films on two dimensional surfaces in addition to hierarchical self-assembly. We present that the DNA-functionalized red-emitting QDs cannot bind to the DNA-functionalized surfaces when they are treated with green laser that are absorbed by these QDs. Further experiments with silica particles excited by a green laser followed by the exposure of red QDs with a red laser reveal that the mechanism enabling the control of binding-unbinding process is the local heating of the nanoparticle surroundings with the light source.

2 Results and Discussion

We first coated the glass surface with ssDNAs and added red-emitting QDs possessing complementary DNAs onto this substrate. During the coating process, we illuminated the substrate using a green laser whose light can be absorbed by the QDs. The section of the sample that was exposed by the laser light appeared as a dark spot on the fluorescence microscopy image, but the area that was not illuminated by the laser light was covered with QDs (Fig. 1).

Next, we studied the binding and unbinding process of the red-emitting QDs on glass substrate while treating the sample using a red laser. Since the red laser light could not be absorbed by the QDs, we observed a homogeneous coating of the QDs on the DNA-functionalized substrate. In another control experiment, we showed that DNA-functionalized silica nanoparticles are not affected by the green laser and successfully coats the surface despite the
laser irradiation. These results clearly show that using light that can be absorbed by the nanoparticles we can locally control the binding-unbinding process of nanoparticles on 2D surfaces.

To study the potential of hierarchical fabrication, we added green-emitting QDs onto the patterned sample prepared using red-emitting QDs and green laser. We observed that the previously uncoated region was successfully coated with green-emitting QDs indicating the possibility of hierarchical self-assembly using our technique.

3 Conclusions

Our results show that DNA-driven self-assembly of nanoparticles on 2D surfaces can be externally controlled using light exposure. We revealed that when the nanoparticles absorb the light, they heat the medium in their close proximity and avoid the binding process of DNA-functionalized nanoparticles on DNA-functionalized surfaces.

Our findings, we believe, will make it possible to create a unique microfabrication technique that relies on the management of DNA-driven nanoparticle self-assembly. We think that this unique technology has the potential to emerge as a viable and affordable replacement for the current fabrication equipment.

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References


Fig. 1. (Left) Illustration of the experimental setup. (Right) Fluorescence microscopy image demonstrating that the spatial control of DNA-functionalized red-emitting QD self-assembly is achieved by employing a green laser.