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Manipulation of Single DNA Using a Micronanobubble Formed by Local Laser Heating on a Au-coated Surface

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We report a method for manipulating sequential single DNA strands by using a micronanobubble formed by local laser heating on a Au surface. DNA strands near the laser focal point were quickly pulled toward the focal point. This DNA pull-in phenomenon can be explained by Marangoni convection due to a micronanobubble generated by laser heating on the Au surface. The thickness of Au film plays a crucial role for absorbing the IR laser light.

DNA engineering, including the use of DNA microarrays,[1] DNA computers,[2] DNA origami,[3] and DNA-templated functional nanomaterials,[4–6] is an important area of research. DNA nanowires are of particular interest because they directly contribute to the fabrication of not only DNA biochips but also the DNA–semiconductor hybrid materials used for scaffolded DNA. The high anionic charge of the DNA backbone enables the bindings of cationic metal ions to be seeded for fabrication of metallic or semiconducting nanowires.[7–9] Since DNA under physiological conditions is generally folded, the extension of DNA on a surface is a prerequisite for biophysical application.

Several methods for extending DNA on a surface have been reported, including electrophoresis,[10] transfer printing on polydimethylsiloxane (PDMS),[11] and molecular combing.[12,13] Among them, molecular combing using air–water interfacial tension is widely used for fabricating DNA-templated nanowires such as metallized wires,[7,14] polymer-coated wires,[15] and magnetic wires.[16] Electrophoresis is used only for DNA extension between two electrodes, and transfer printing on a PDMS sheet requires that the number of DNA be controlled. There are other possible methods besides these for manipulating single DNA.

We have developed a method based on the use of a Nd:YAG laser for manipulating DNA strands sequentially at a particular position and for controlling the number of DNA on the surface. A micronanobubble is formed on a Au thin film by local laser heating, which induces Marangoni convection around the bubble.[17] The resulting Marangoni convection produces the pull-in effect common to nanomaterials such as DNA. With this method, we can for the first time manipulate single DNA on a Au-coated surface at the laser focal point. Even though we used a Nd:YAG laser for our experiments, the principle of the manipulation differs significantly from that of laser trapping with optical tweezers.

Our new observation using this DNA manipulation method was reversible pull-in and shrinkage of one end of immobilized double-stranded DNAs when the Nd:YAG laser was switched on and off. A custom fluid cell (Figure 1a) was constructed using two plates of Au-coated quartz and a cover glass separated by two Parafilm™ spacers with a thickness of 40 μm. The cell was

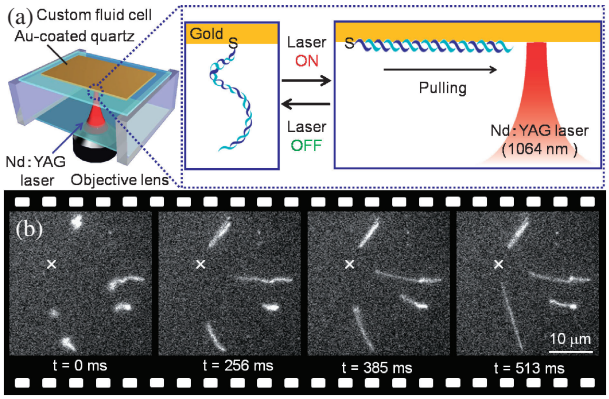

Figure 1. (a) Schematic representation of custom fluid cell mounted on objective lens and DNA response to laser irradiation. (b) Fluorescent images of temporal changes in DNA stands. Strands were pulled in direction of laser focal point (indicated by ×).

mounted on an *xyz* translational cell positioner. Thiol-labeled λ-DNA was used, and the DNA strands were directly immobilized by dipping the Au substrate into a solution containing the DNA (7.0×10^{-16} M) for 10 min. The DNA strands were sparsely attached through the thiol group at one end, and the opposite end remained free on the Au surface. λ-DNA visualized by staining with SYBR Gold™ was introduced into the fluid cell by capillary force. The drifted Brownian motion of micrometer-sized globes was observed by fluorescence microscopy using 460–495-nm illumination. The globes were composed of the stained self-coiled DNA in the solution. Then a continuous YAG laser beam (1064 nm) was focused on the Au surface through an oil-immersion objective lens (NA = 1.3). When the focal point of the laser beam was adjusted to the top of the Au surface, the self-coiled DNA strands were pulled rapidly toward that point (Figure 1b). When the laser irradiation was stopped, the expanded strands returned to their original state (Figure S1).[18] Switching the laser on and off caused the DNA to pull-in and shrink reversibly many times.[19,20]

This pulling behavior was not observed when the laser was focused on a glass or quartz surface. On a Au-patterned quartz plate, the pulling suddenly stopped when the focal point was moved from the Au surface to that of the SiO_2. Furthermore, the pull-in effect was sensitive to the position of the focal point normal to the surface. When the focal point was moved vertically from the top of the Au surface to the bulk solution (i.e., it left the surface), this pull-in phenomenon was not observed. Therefore, this phenomenon cannot be explained by using the same principle as that of laser trapping with optical tweezers.[21] In addition, the laser power and Au film thickness

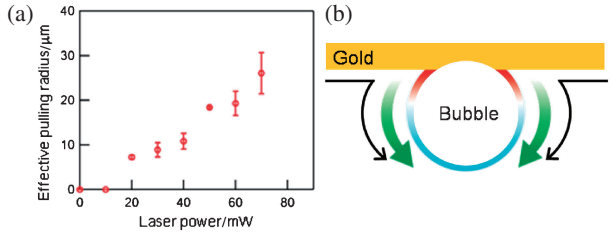

Figure 2. (a) Dependence of pulling radius on laser power for 10-nm-thick Au film. (b) Schematic showing flow of Marangoni convection inducing laser pull-in effect around micronanobubble on Au surface locally heated. Black arrow is water flow. Green arrow is surface tension gradient. Red and blue curve lines indicate temperature gradient of liquid–gas interface. Red and blue show high and low temperature.

affected the pull-in phenomenon. A minimum laser power of 20 mW was needed for the pull-in phenomenon to occur when the film was 10 nm thick. As the laser power was increased beyond 20 mW, the effective pulling radius, which is defined as the radius of a concentric circle around the focal point and the location of the immobilized strands, increased (Figure 2a). Increasing the Au film thickness increased the required laser power for DNA pulling. With 10-nm-thick Au film, 34% of the infrared (IR) light irradiation at 1064 nm was absorbed, 41% was transmitted, and 25% was reflected. However, the IR absorption was substantially lower for thicker Au film: 28 and 5% for 20 and 50 nm thicknesses, respectively. Moreover, thermal conductivity increased with the thickness.[22] Therefore, the IR light absorption of the Au film plays an important role in DNA pulling, which is supported by the our finding that the pulling effect occurred not only for a Au surface but also for a glassy carbon surface and a glass surface written on with a black marker pen. The present DNA motion at the laser focal point on Au thin film is not caused by thermophoresis (Soret effect),[23] since the thermophoresis effect leads to a divergent fluidic movement around the laser focus, resulting in DNA elongation along the thermal gradient with relatively slow DNA movement (less than $1\,\mu m\,s^{-1}$). In contrast, the present DNA motion induced by focusing a laser beam on Au film was too fast ($>40\,\mu m\,s^{-1}$) to be accounted for by a thermophoresis mechanism.

The existence of a laser power threshold strongly indicates the presence of a phase transition. The formation of a micronanobubble at the Au/water interface due to local heating is the most probable mechanism for the DNA pulling. The direct observation of a micronanobubble is not so easy at a low laser power of 20 mW, but an opaque sphere-shaped object was observed at a higher laser power (>50 mW) with a fluorescent microscope. The local heating at the laser focal point caused water vaporization at the Au/water interface, resulting in the formation of a micrometer-sized bubble at the laser focus. In this micronanobubble, the thermal gradient induced surface tension gradients at the bubble–water interface since the surface tension was higher at lower temperature, which is the driving force of Marangoni fluidic convection into the laser focal point (Figure 2b).[17]

In summary, we have observed, for the first time, reversible pull-in and shrinkage of DNA strands near a Nd:YAG laser focal point on a Au surface. This phenomenon can be explained by Marangoni convection at the micronanobubble/water/Au interface. The present study is applicable to single DNA strand manipulation. This optical method opens the door to the manipulation of single DNA for biophysical application.

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