Supplemental Material

for

Light-induced reversible expansion of individual gold nanoplates

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Contents

1 Microfiber fabrication and micrometer-sized gold plate synthesis

The microfiber is fabricated using a flame-heated drawing technique [1]. The fabrication procedures can be described as follows: A certain region of coating layer of a single mode optical fiber is removed. Put the prefabricated fiber on (hydrogen flame torch) and pull the fiber slowly by the two motorized translation stages. The diameter of the microfiber is determined by the pulling distance, which ranges from several hundred nanometers to several micrometers. The gold plate used here is synthesized in a mixture of chloroauric acid ($AuCl_3 \cdot HCl \cdot 4H_2O$) and aniline ($C_6H_7N$) as reported previously [2]. The side length and thickness of synthesized gold plate is several micrometer to a dozen micrometers and dozens of nanometers, respectively.

2 Photothermal expansion measurements

2.1 Experimental procedures

Firstly, The gold plates are deposited on the glass substrate and then dried. The tapered fiber is manipulated by the three dimensional console to pick up the gold plate indicated in Fig. S1(a) and transfer it to the microfiber indicated in Fig. S1(b). Secondly, the gold plate-microfiber system is put into the SEM chamber and the microfiber is connected with a fiber laser (wavelength, 980 nm) by an optical-fiber-through-vacuum connector. Finally, turn on the laser and monitor the changes of the gold plate on the microfiber with electron microscope at the same time.

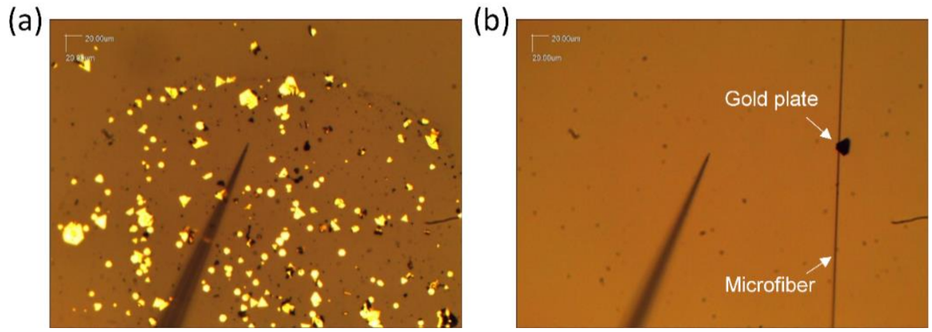

Figure S1 | Transferring a gold plate from glass substrate to the microfiber.

2.2 Expansion measurements

The photothermal expansion of the gold plate after the laser guided into the microfiber can be measured with the help of the electron microscope. As Figure S2 shows, under different laser power, we take two SEM images of the gold plate before and after the laser switched on. Since everything (including imaging settings of the SEM, position of the gold plate, etc) except the ON-OFF state of laser are the same for these two SEM pictures, we can compare the area of the gold plate before and after laser switched on and obtain the expansion. For convenience, we select three well-defined points on the gold plate and calculate the area of the triangle defined by these three points. The pixel coordinates of these three points can be obtained from the SEM picture. We can convert the pixel length to the real length μm according to the scale bar in these images. Then we can obtain the results as Table S1 shows.

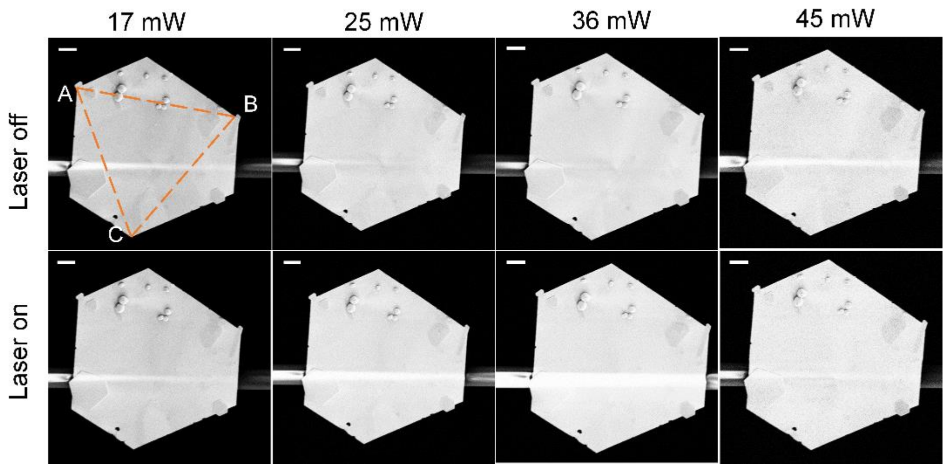

Figure S2 | SEM images under different laser power when laser is off and on. The red dashed triangle defined by three well-selected points (A, B, C) on the gold plate in is used to measure the area before and after expansion. Scale bars are 2 μm.

P [mW]	Laser	A [pix]	B [pix]	C [pix]	Area [μm^2]	ΔA/A [%]
17	off	(311,154)	(807,243)	(481,619)	133.60	0.93
	on	(344,151)	(842,238)	(517,618)	134.84	
25	off	(335,154)	(811,239)	(500,599)	133.62	1.40
	on	(369,151)	(848,235)	(536,599)	135.49	
36	off	(343,146)	(861,240)	(524,628)	133.67	2.16
	on	(382,141)	(905,234)	(565,628)	136.56	
45	off	(330,138)	(848,230)	(506,618)	133.55	2.94
	on	(359,129)	(882,221)	(538,618)	137.48	

Table S1 | Measured pixel coordinates of the three corners (A, B, C) in the triangle, the area of the triangle, and the relative area expansion (ΔA/A) under different laser power.

In order to obtain the exact photo-thermo-mechanical response time of the gold nanoplates, the time for laser power to increase from zero to the setting value when the laser is switched on (also, the time for laser power to decrease from the setting value to zero when the laser is switched off) should be measured. As Figure S3 shows, it takes 0.17 s for the laser power to increase to the setting value and 0.35 s for the laser power to decrease to zero when the laser is switched on and off.

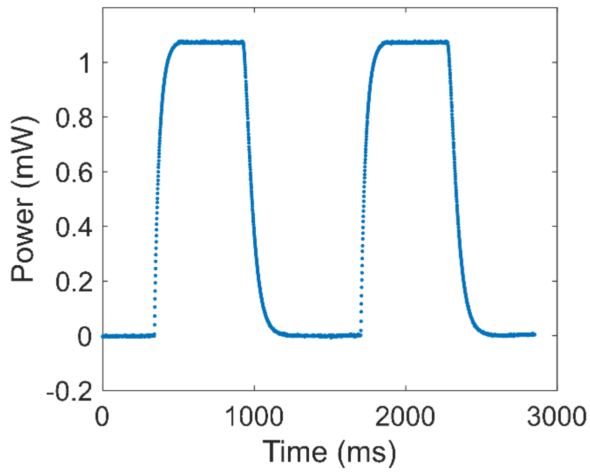

Figure S3 | Laser power response time when laser is switched on and off in succession. It takes 0.17 s for the laser power to increase to the setting value and 0.35 s for the laser power to decrease to zero when the laser is switched on and off, respectively.

3 Temperature calculations

3.1 Electromagnetic field simulations

Here, we use the commercial software *FDTD solutions* (v8.13, Lumerical) to calculate the electric and magnetic field distributions with 3D simulations. We use a grid size (dx, 100 nm; dy, 1 nm; dz, 100 nm) for the gold plate-microfiber structure. We use mode source in FDTD software to obtain the fundamental mode of the microfiber for wavelength of 980 nm. In order to appropriately reduce both the simulation memory and simulation time, we have tuned the grid size in simulation and find this value is enough in the accuracy.

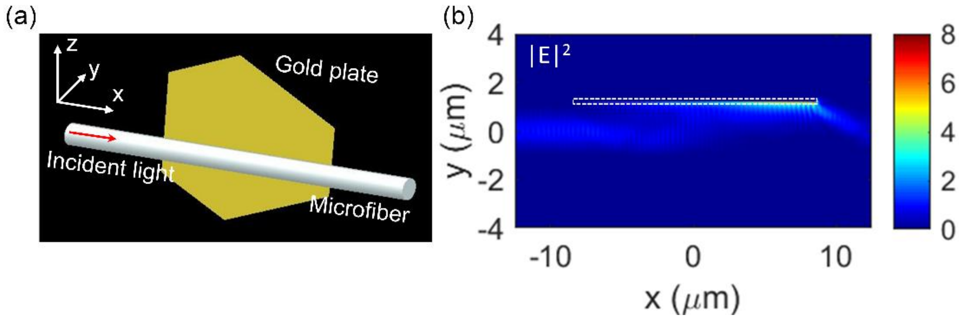

Figure S4 | Electromagnetic field simulation. (a) Schematic of the gold plate-microfiber system. (b) Electric field distribution (XY plane through the center of the microfiber).

3.2 Temperature simulations

From the FDTD results, the electric field distribution in the gold plate can be extracted and the heat power volume density $Q_d(x, y, z, \lambda)$ can be calculated as [3]

$$Q_d(x, y, z, \lambda) = \frac{1}{2}\varepsilon_0 \omega Im(\varepsilon_r)|\mathbf{E}(\mathrm{x}, \mathrm{y}, \mathrm{z}, \lambda)|^2 \tag{S1}$$

The temperature distribution of the gold plate-microfiber system is then calculated with COMSOL Multiphysics using the calculated $Q_d(x, y, z)$ as input. The thermal conductivity of the gold plate with thickness 30 nm is taken as [4] 150 W/(m·K).

In order to get the exact temperature of the gold plate, the laser power in the microfiber where gold plate is placed on should be obtained. However, under our experiment condition, we can only measure the laser power at the input port of the microfiber. Since some power may disappear on the way to the gold plate so we need to know the attenuation from the input port to the gold plate. This is done in two steps.

First we find the melting point for the micro-sized gold plate. As Figure S5 shows, the gold plate is heated by the hot stage under different temperature for 1 minute and then placed in the SEM chamber for observation. The gold plate starts edge-melting when temperature is near 450 °C and center-melting when temperature is near 550 ℃. When the temperature is increased to 600 ℃ (see Figure 3(d) in the main text), most of the gold plate is melted, which indicates that the melting point of the gold plate can be considered as around 600 ℃.

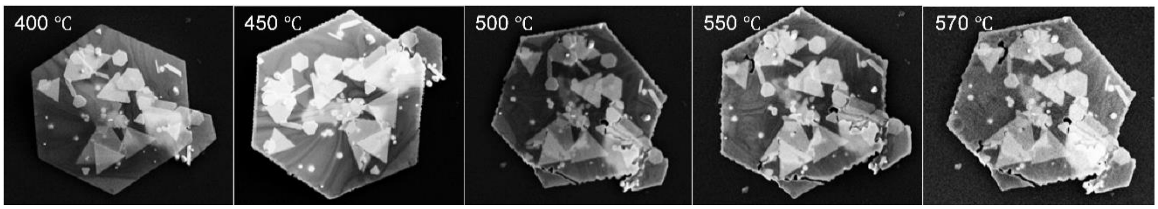

Figure S5 | Morphology evolution of the gold plate heated by hot stage with temperature increasing. The gold plate starts edge-melting when temperature is near 450 ℃ and center-melting when temperature is near 550 ℃.

We then take our sample for which we have calculated the relative area expansion and put it in the SEM. The power through the fiber is increased until the gold plate melts noticeably. This happens at 57 mW (see Figure 3(c) in the main text). The new gold plate is then compared with the heated sample earlier to decide what temperature this corresponds to. It is reasonable to assume that using a power of 57 mW will give us a temperature around 600 ℃.

In the second step we use the aforementioned way to simulate the effective power at the gold plate corresponding to this melting temperature. The effective power is 8.6 mW. The attenuation should be 8.6/57. So for the input power as 17, 25, 36, 45, 57 mW, the effective power at the gold plate should be 2.56, 3.77, 5.43, 6.79, 8.6 mW, respectively. We use the effective power as input in our simulations to get the temperature of the gold plate.

4 Error analysis

The error mainly comes from two aspects. One is the resolution limitation of the electron microscope. Even though the resolution can reach several nanometer for our electron microscope, the relative resolution will be reduced when we measure a large object (i.e. the gold plate with size of a dozen microns). This will cause error in the expansion area calculation. Here we consider the error of the pixel coordinate to be ±2 pixels, then we can obtain the relative area expansion ($\Delta A/A$) as 0.93%±0.14%, 1.40%±0.15%, 2.16%±0.15%, 2.94%±0.15% when the laser power is 17, 25, 36, 45 mW, respectively.

The other is the melting range of the gold plate, causing the error in determining the melting point of the gold plate and in the temperature calculation. Here we consider the error of the melting point of the gold plate to be ±50 ℃. Then the calculated temperature is 240.6±14.9, 312.5±22.0, 411.3±31.6, 492.2±39.5, 600±50 ℃ when the laser power is 17, 25, 36, 45, 57 mW, respectively. To calculate error of the photothermal expansion coefficient α, we can use $\alpha_{\pm} = \frac{(\Delta A/A)_{\pm}}{2\Delta T_{\mp}}$. Finally, we obtain the expansion coefficient as 21.4±4.6, 24.3±4.4, 27.9±4.1, 31.5±4.2 $\mu\cdot K^{-1}$ when the laser power is 17, 25, 36, 45 mW, respectively (i.e. when the temperature is 240.6±14.9, 312.5±22.0, 411.3±31.6, 492.2±39.5 ℃, respectively).

5 Difference between this work and our previous PRL work

We would like to provide a clear demonstration of the difference between this work and our previous PRL work (Phys. Rev. Lett. 118, 043601 (2017)). In the previous PRL paper, a pulsed laser is used to study movements of gold plates on a tapered fiber due to synergic effect of photophoretic force and optical force in air, whereas in this work, a CW laser is used to study the thermal expansion of gold plates on a uniform microfiber due to photothermal effect in vacuum. Therefore, the experimental setups, the underlying physical processes, and the results are completely different.

Our previous PRL work

Our previous PRL work demonstrates a way to drive a gold plate to move along a tapered fiber guided with supercontinuum light (pulsed) in air mainly by the photophoretic force, as well as with the optical force. An introduction to the physical process is stated below. For the tapered fiber, the diameter of the fiber becomes smaller towards the fiber tip, and the evanescent field outside of the fiber becomes stronger especially at the tapered fiber tip, making a different light-induced heat source distribution in the gold plate. Therefore, a small temperature gradient along the gold plate is formed. Air molecules striking the plate with different speeds along the temperature gradient, which produces a net force (i.e. photophoretic force) causing translational movement of the gold plate. The thermal expansion of the gold plate was not considered as it probably does not contribute to the movement of the gold plate.

This work

In this work we investigated the thermal expansion of the gold plate based on a different experimental setup. We use a microfiber with uniform diameter, and the fiber is guided with continuous wave (CW) laser (wavelength 980 nm). The gold plate is stuck on the microfiber and does not move. The setup is in vacuum (SEM chamber). In this case, the air density is very low which results in negligible photophoretic force (which plays a major role shown in our PRL work). The physical processes happening in this case are described as follows: Laser light is absorbed by the gold plate, which heats up the gold plate. The gold plate expands due to this temperature increase. When the laser is switched off (i.e., the heat source is removed), the gold plate will contract back naturally as the sample cools down.

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