Supporting Information

Remotely controlled colloidal assembly of soft microrobotic artificial muscle

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Supplementary Notes

The energy density absorbed by an artificial µmuscle, $E_{\mu\text{muscle}}$, was estimated using the following equation. We assume that all the energy carried by the photons that reach the surface of the gold nanoparticle is converted into thermal energy. This formulation was deduced from Beer-Lambert’s law\[^{[32]}\].

$$E_{\mu\text{muscle}} = \frac{P_{\text{laser}}(1 - \xi)^2(1 - 10^{-e_{\text{AuNR}}.l}.c} {\sigma.l}$$

where $P_{\text{laser}}$ is the laser power measured on the glass slide, $\xi$ is the scattering coefficient of IR window, $e_{\text{AuNR}}$ is the molar absorption coefficient of the AuNR, $l$ is the optical pathlength, $c$ is the molar concentration of the gold nanorods in the artificial µmuscle, $t$ is the laser exposure time, and $\sigma$ is the area of the laser beam.

Note that, for simplification, the contributions of the Pt and Ni layers were not considered. The molar concentration of particles in an artificial µmuscle was estimated with the following equation:

$$c = \frac{1}{V_{m\text{NA}}.N_A}$$

where $V_{m\text{NA}}$ is the volume of a single mNA and $N_A$ is the Avogadro number.
The values of the parameters in our system are as follows: $P_{\text{Laser}} = 14.22$ mW, $\varepsilon_{\text{AuNR}} = 4.57 \times 10^9$ $\text{M}^{-1} \text{cm}^{-1}$ at 785 nm, $\xi = 0.04$, $l = 5 \mu\text{m}$, $\sigma = 1.96 \times 10^{-9}$ $\text{m}^2$, and $c = 4.96 \times 10^{-8}$ M. For the chosen parameters, $E_{\mu\text{muscle}} = 3.06 \times 10^{11}$ J·m$^{-3}$. 
Supplementary Figures

Figure S1 (a) Representative STEM-EDX line scan obtained for the Au/Pt/Ni+pNIPMAM nanoparticle shown in Figure 1. Au (green), Pt (red) and Ni (dark blue). Carbon (cyan) was added as a control. (b) HAADF (High-angle annular dark-field) image showing the different layers of metals (left), and bright-field image showing the direction of the line scan. Scale bar, 50 nm.

Figure S2 Representative transmission electron microscopy (TEM) images of the gold nanorods at (a) low and (b) high magnification. The average size of the nanorods was calculated using such microscope images. Scale bars, 50 nm.
Figure S3 Experimental platform. (a) A schematic illustration showing key components of the system. The NIR illumination is provided by a laser that is connected to the fluorescence recovery after photobleaching (FRAP) module of the microscope via fiber optic cable. A Digital Micromirror Device (DMD) projects UV light for the photopolymerization of the hydrogel structures. The motorized inverted microscope has two turrets stacked on top of each other to enable simultaneous laser illumination and fluorescence imaging. SOLA light source provides the illumination required for fluorescence imaging of tracer particles. The environmental chamber around the system provides controlled temperature and humidity during the experiments. The magnetic control system was mounted on top of the stage around the sample. Rectangles colored cyan show the optical path. (b) A picture of the experimental platform.
Supplementary Movies

**Movie S1** Reversible assembly of mNAs under a homogeneous rotating magnetic field (H = 40 mT). The particles form chains at 0.1 Hz which become smaller and fragmented at 1 Hz. The video is played in real time.

**Movie S2** Representative example showing *in situ* photothermal colloidal assembly of artificial µmuscle. The video is played in real time.

**Movie S3** Thermocapillary flows visualized with fluorescent particles at a laser power of 11 mW. The video is played in real time.

**Movie S4** Optical actuation and frequency modulation of a spherical mMA. It is shown the contraction and relaxation of the assembly for different actuation frequencies: single twitch (with illumination time of 1000ms), 0.5, 4 and 8Hz (with 100ms of illumination). The laser power density used was 7μW/μm². For the single twitch and 0.5Hz, the mMA recovers its original shape. The video is played in real time.

**Movie S5** Laser writing of a square-shaped artificial µmuscle. The video is played in real time.

**Movie S6** Magnetic actuation of a triangle-shaped artificial µmuscle using homogeneous rotating magnetic fields (H = 40 mT) at different frequencies. The video is played in real time.

**Movie S7** Controlling the position of an artificial µmuscle using a single magnet. The video is played in real time.

**Movie S8** Reversible photothermally-driven agglomeration of mNA that are not functionalized with amine groups. When the laser is switched on, mNAs concentrate at the illumination area. As soon as the laser is switched off, the particles disperse, showing that the particles fail to permanently attach to each other. The video is played in real time.

**Movie S9** Assembly of artificial µmuscle from mNAs functionalized with alkyne groups instead of amine groups. When the laser is switched off the particles do not disperse and stay attached to each other, verifying that permanent in situ colloidal assembly works with alkyne groups. The video is played in real time.

**Movie S10** Photothermal assembly of artificial µmuscle between the arms of a lever mechanism and optical actuation of the micromachine. The video is played in real time.