Supplementary Materials for

Photopyroelectric microfluidics

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**Note S1. Design guidelines**

To initiate droplets’ motions, the dielectrophoretic force $F_e \propto (P_c \omega_1 \omega_2 \alpha P)^2$ should overcome the adhesive force $F_\gamma$, where $\omega_1$ is the transmittance of superomniphobic surface, $\omega_2$ and $P_c$ are the transmittance and pyroelectric coefficient of the pyroelectric crystal, respectively, $\alpha$ is the photothermal conversion coefficient of the photothermal film, $P$ is the laser power. This requires properties of three functional layers as follows:

For the superomniphobic surface, the adhesive force $F_\gamma$ should be low and the transmittance $\omega_1$ should be high to allow ready light transmission. The thickness of the surface $d_s$ should be small enough so that the electric field is sufficiently strong ($d_s < 1 \text{ mm}$). The high transparency is also required for the pyroelectric crystal (transmittance $\omega_2$) as it is placed atop the photothermal film. Moreover, the crystal should have strong pyroelectricity $P_c$ to effectively convert the thermogenesis into extra surface charges. For the photothermal film, it should have high photothermal conversion coefficient $\alpha$ to generate heat upon light irradiation and low thermal conductivity to reduce heat loss.

**Note S2. Analysis of heat transfer within photothermal film**

Take the photothermal film as a thin plate. The Bi number, the ratio between heat conduction resistance and surface heat convection resistance, is expressed as follows:

$$\text{Bi} = \frac{hd}{\lambda}$$

where $d$, $h$, and $\lambda$ denotes the thickness of photothermal film, convection heat transfer coefficient, and thermal conductivity, respectively.

With $d = 1 \times 10^{-4} \text{ m}$, $h = 5 \text{ W m}^{-2} \text{ K}^{-1}$, $\lambda = 0.5 \text{ W m}^{-1} \text{ K}^{-1}$, we have $\text{Bi} = 0.001$. Therefore, the temperature $T$ can be approximated to be uniform in $z$ direction and only varies in $r$ direction (Fig. S9).

As a result, the heat transfer in photothermal film can be simplified to be heat conduction in a semi-infinite cylinder. The mathematical problem can be described as follows:

$$\frac{1}{a} \frac{\partial T}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right), \quad 0 < r < \infty$$

$$t = 0, \; T(r, t) = T_0$$

$$r = R, \; -\lambda \frac{\partial T}{\partial r} = q_0$$
where \( a, t, r, R, T_0 \) and \( q_0 \) denotes thermal diffusivity of photothermal film, time after laser irradiation, distance to laser, laser spot radius, ambient temperature, and heat flux, respectively. The heat flux is determined by \( q_0 = P/(2\pi R d) \). The analytical solution of the problem can be readily obtained as follows:

\[
T(r,t) = T_0 + \frac{2q_0 R}{\pi \lambda} I
\]

\[
I = \int_0^\infty \frac{1-\exp\left(-\frac{atu}{R^2}\right)}{a^2} \times \frac{Y_l(u)J_0\left(\frac{r}{R}u\right)-J_l(u)Y_0\left(\frac{r}{R}u\right)}{J_1(u) + Y_1(u)} \, du
\]

The maximum temperature \( T_{\text{max}} \) occurs at \( r = R \) and is expressed as follows:

\[
T_{\text{max}} = T_0 + \frac{2q_0 R}{\pi \lambda} I(R,t)
\]

\[
I(R,t) = \int_0^\infty \frac{1-\exp\left(-\frac{atu}{R^2}\right)}{a^2} \times \frac{Y_l(u)J_0\left(\frac{r}{R}u\right)-J_l(u)Y_0\left(\frac{r}{R}u\right)}{J_1(u) + Y_1(u)} \, du
\]

The analytical solution shows that the temperature rise \( \Delta T = T_{\text{max}} - T_0 \) is proportional to heating power \( P = 2\pi R q_0 d \). Using the Hasan equation, the solution can be further simplified as follows:

\[
T_D = \begin{cases} 
1.1281\sqrt{\tau_D}\left(1 - 0.3\sqrt{\tau_D}\right), & \tau_D \leq 1.5 \\
(0.4063 + 0.5\ln \tau_D)\left(1 + \frac{0.6}{\tau_D}\right), & \tau_D > 1.5 
\end{cases}
\]

\[
\tau_D = \frac{t}{R^2}
\]

\[
T_D = (T_{\text{max}} - T_0) \frac{\lambda}{q_0 R}
\]

\[
\tau_D = \frac{at}{R^2}
\]

**Note S3. Numerical study of temperature and electric field distributions**

The temperature distribution within the lithium niobate crystal is firstly studied using a finite element method. The heat conduction equation is as follows:

\[
\rho c \frac{\partial T}{\partial t} - \nabla \cdot (\lambda \nabla T) = s(r,t)
\]

where \( \rho, c \) and \( s \) denote the mass density, specific heat and heat source, respectively.

The Neumann-type boundary condition reads:

\[
\hat{n} \cdot \lambda \nabla T = h(T_0 - T)
\]
where \( \vec{n} \) is the inward normal to the boundary.

The initial condition is as follows:

\[
t = 0, \quad T = 298.15 \text{ K}
\]

As the lithium niobate crystal is heated by the underlying photothermal film through laser irradiation, the heat source term is expressed as follows:

\[
s(r,t) = \frac{P \omega_1 \omega_2 \alpha}{\pi R^2 d_p} (0 \leq r \leq R)
\]

\[
s(r,t) = 0 (r > R)
\]

where \( d_p \) denotes the thickness of lithium niobate crystal. In our case, \( \omega_1 = 0.68, \omega_2 = 0.72, \alpha = 0.99, R = 1.0 \text{ mm and } d_p = 0.5 \text{ mm.} \) The resultant temperature mapping within the lithium niobate after the laser is turned on for 1 s is shown in Fig. S10.

Similarly, the electric field distribution is simulated using a finite element method. The governing equations are expressed as follows:

\[
\vec{E} = -\nabla V
\]

\[
\nabla \cdot (\varepsilon_r \varepsilon_0 \vec{E}) = \rho_i
\]

where \( \vec{E} \) is the strength of electric field, \( V \) is the electric potential, \( \varepsilon_0 \) is the vacuum permittivity, \( \varepsilon_r \) is relative permittivity, and \( \rho_i \) is charge density.

Charge density boundary conditions are given on the lithium niobate wafer surface as follows:

\[
\vec{n}(\vec{D}_i - \vec{D}_z) = P_i (T - T_0)
\]

where \( \vec{D}_i \) and \( \vec{D}_z \) are the electric displacement of lithium niobate crystal and air at the interface, respectively. The above superomniphobic surface is very thin (~200 µm), its presence has negligible impact on the electric field distribution (Fig. S11).

The crystal is initially at ambient temperature. Its spontaneous polarization is compensated by free charges. Thereby, the initial condition for electric potential is given as follows (32):

\[
V = 0
\]

**Note S4. Thermocapillary effect**

On a solid surface with a temperature gradient \( \nabla T \), droplets experience thermocapillary forces owing to two effects: (a) minimization of surface energy to decrease the surface energy (capillary forces drive droplets towards areas of larger spreading coefficient \( S = \gamma_{sa} - (\gamma_{ai} + \gamma) = \gamma(\cos \theta - 1) \))
, where subscript sa and sl denote solid/air and solid/liquid interface); (b) Marangoni flows caused by the surface tension gradients $\nabla \gamma$ on liquid/air interface (39).

By simplifying the system into one-dimensional, the thermocapillary force $f_{th}$ can be written as follows (40):

$$f_{th} = l \left( \frac{dS}{dT} + \frac{d\gamma}{dT} \right) \frac{dT}{dr}$$

where $l$ is the liquid/solid contact length. By measuring the spatial distribution of temperature on the superomniphobic surface upon laser irradiation, we obtain the maximum temperature gradient $dT/dr = 2.5 \, ^\circ C \, mm^{-1}$ (Fig. S7). For a 5 $\mu$l water droplet, the contact length on the superomniphobic surface is $l = 0.83 \, mm$. At 20-30 $^\circ C$, the water surface tension $d\gamma/dT \approx -0.15 \, mN \, m^{-1} \cdot ^\circ C^{-1}$ and the contact angle $\theta$ barely changes. Thus, we can have $f_{th} \approx 0.305 \, mN \cdot m^{-1}$. By multiplying $f_{th}$ with the radius of droplets, the magnitude of the force is roughly 0.305 $\mu$N which is two orders of magnitude lower than the dielectrophoretic force.

**Note S5. Analytical model for droplet’s motion onset**

When the droplet is far away from the laser ($> 5 \, mm$), we approximate the surface charges on the irradiated lithium niobate wafer to be a point charge. The electric field strength induced by the point charge $Q$ is thus expressed as follows:

$$E = K_e \frac{Q}{r^2}$$

where $K_e$ is Coulomb constant ($9 \times 10^9 \, N \, m^2 \, C^{-2}$).

The dielectrophoretic force exerted on the droplet by the point charge is expressed as follows:

$$F_e = 4\pi r_0^3 \varepsilon_0 k (E \cdot \nabla) E$$

where $r_0$ is the droplet radius, $k$ is the Clausius-Mossotti factor ($k = (\varepsilon - \varepsilon_0)/(\varepsilon + 2\varepsilon_0)$), $\varepsilon_0$ and $\varepsilon$ are the permittivity of air and droplets, respectively.

The dielectrophoretic force can be rewritten as follow:

$$F_e = -8\pi r_0^3 \varepsilon_0 k \frac{K_e^2 Q^2}{r^5} \propto \frac{kQ^2}{r^5}$$

As the surface charge $Q$ is related to pyroelectric coefficient ($P_e = -8.3 \times 10^{-5} \, C \, m^{-2} \cdot ^\circ C^{-1}$) by (32),

$$Q = P_e A \Delta T$$
where $A$ is the area of laser irradiated region, and $\Delta T$ is the temperature rise of lithium niobate wafer. As mentioned in Note S2, the temperature rise $\Delta T$ is proportional to the laser power $P$:

$$\Delta T \propto P$$

Thus, the dielectrophoretic force can be described as follows:

$$F_E \propto \frac{kP^2}{r^5}$$

When the droplet positioned at a distance $r$ starts to move, its adhesive force $F_\gamma$ is overcome by dielectrophoretic force $F_E$ so that $F_E = F_\gamma$, where $F_\gamma \propto r^{-1}$ is concluded by fitting experimental data. Thus, the radius $r_{\text{max}}$ of the trapping domain which is the maximum distance at which the droplet could be attracted to the laser, is derived as follows:

$$r_{\text{max}}^5 \propto k_\gamma P^2$$
**Fig. S1.** High-resolution transmission electron microscopy image of a dendrite of the superomniphobic surface networks, showing the slightly sintered hollow silica nanospheres. Inset is the diffraction pattern which shows the amorphous phase of silica.
Fig. S2. Roughness of the superomniphobic surface. (A) Scanning electron microscopy image of the superomniphobic surface. (B) Height profile measured using a laser profilometer. (C) Surface roughness profile along the marked line in (B). (D) Roughness distribution.
Fig. S3. Transmittance of superomniphobic surfaces to visible and near infrared light.
Fig. S4. Contact angles of diverse droplets deposited on the superomniphobic surface with angle values listed in Table S1.
Fig. S5. Schematic showing the forces exerting on the droplet. Initially, the lateral $F_{E,r}$ counteracts the resistant force $F_{r}$, accelerating the droplet. As the droplet approaches the laser, the $F_{E,r}$ reverses its direction and decelerates the droplet. Vertically, the substrate supporting force $F_s$ balances the vertical component of dielectrophoretic force $F_{E,z}$ and droplet gravitational force $G$. 
Fig. S6. Sensitivity of the results to the position of extracted lines. (A) Mapping of $E_r(\partial E_r/\partial r)$ surrounding the heated pyroelectric crystal. (B) The spatial profile of $E_r(\partial E_r/\partial r)$ along the dashed extracted lines in (A). (C) The variation of peak values of $E_r(\partial E_r/\partial r)$ with the position $z$ of extracted lines in (A).
**Fig. S7. Temperature distribution on the superomniphobic coating.** (A) Infrared thermal imaging showing the cross-sectional temperature distribution upon 400 mW laser irradiation for 60 s. (B) The spatial distribution of temperature and temperature gradient on the superomniphobic surface in (A).
Fig. S8. Confocal microscopy images of protein droplets’ moving trails. (A) Fluorescence showing the protein absorption on the digital microfluidic platform. Reprinted with permission from Ref. (31), Copyright (2008) American Chemical Society. (B) No fluorescence can be observed on the photopyroelectric platform.
Fig. S9. Temperature distribution within photothermal film.
Fig. S10. Temperature distribution within the lithium niobate wafer at \( t=1 \) s.
Fig. S11. Impact of the superomniphobic surface on the electric field distribution. Mapping of $E_z(\partial E_z/\partial r)$ as the thickness $d_s$ of superomniphobic surface is (A) 0.1 mm and (B) 0.2 mm. (C) The strength of $E_z(\partial E_z/\partial r)$ slightly decreases with increasing $d_s$. The extracted line is 1 mm above the superomniphobic surface.
Table S1. The static contact angles and roll-off angles of various liquids on the superomniphobic surface.

| Liquids                  | Surface tension (mN m⁻¹) | Dielectric constant | Density (kg m⁻³) | Contact angle (º) | Contact angle hysteresis (º) | Roll-off angle (º) |
|--------------------------|--------------------------|---------------------|------------------|------------------|-----------------------------|-------------------|
| Silicone oil (PDMS)      | 18.9                     | 2.7                 | 963              | 150.6            | 12.4                        | 5±1               |
| n-Heptane                | 20.1                     | 1.9                 | 680              | 151.6            | 11.2                        | 5±1               |
| Isopropyl alcohol (IPA)  | 20.9                     | 18.3                | 786              | 150.3            | 10.9                        | 5±1               |
| n-Octane                 | 21.2                     | 1.9                 | 703              | 151.6            | 10.8                        | 5±1               |
| Ethanol                  | 21.8                     | 24.3                | 789              | 150.5            | 10.3                        | 5±1               |
| Acetone                  | 22.7                     | 20.7                | 785              | 153.0            | 9.2                         | 5±1               |
| Dichloromethane          | 23.2                     | 9.1                 | 1325             | 154.2            | 8.9                         | 4±1               |
| Ethyl acetate            | 23.3                     | 6.0                 | 902              | 155.0            | 8.9                         | 4±1               |
| n-Decane                 | 23.4                     | 2.0                 | 730              | 155.4            | 8.2                         | 4±1               |
| n-Butanol                | 24.0                     | 17.5                | 810              | 152.6            | 8.1                         | 4±1               |
| Cyclohexane              | 24.4                     | 2.1                 | 780              | 155.0            | 7.6                         | 4±1               |
| Dimethyl carbonate       | 24.6                     | 3.1                 | 980              | 154.3            | 7.5                         | 4±1               |
| n-Dodecane               | 25.0                     | 2.0                 | 749              | 155.7            | 7.8                         | 4±1               |
| 1,2-Dichloroethane       | 25.1                     | 10.4                | 1235             | 155.5            | 7.0                         | 4±1               |
| n-Octanol                | 27.3                     | 20.3                | 830              | 153.1            | 6.8                         | 4±1               |
| n-Hexadecane             | 27.5                     | 2.1                 | 773              | 157.2            | 6.8                         | 4±1               |
| Acetic acid              | 27.7                     | 6.2                 | 1050             | 156.5            | 6.3                         | 4±1               |
| Toluene                  | 28.0                     | 2.4                 | 866              | 157.0            | 6.0                         | 4±1               |
| n-Decanol                | 29.8                     | 8.1                 | 830              | 158.9            | 5.3                         | 3±1               |
| Dimethyl formamide       | 35.6                     | 38.2                | 945              | 158.6            | 4.8                         | 3±1               |
| Benzyl ether             | 39.8                     | 3.9                 | 1040             | 160.8            | 4.4                         | 3±1               |
| Concentrated HNO₃, 16 M  | 41.2                     | 19.0                | 1400             | 162.1            | 2.4                         | 2±1               |
| Dimethyl sulfoxide       | 42.7                     | 47.2                | 1100             | 162.9            | 3.5                         | 2±1               |
| Ethylene glycol          | 48.2                     | 37.7                | 1116             | 157.4            | 2.8                         | 2±1               |
| Concentrated H₂SO₄, 18 M | 55.1                     | 8.6                 | 1840             | 163.0            | 2.1                         | 2±1               |
| Glycerol                 | 64.8                     | 42.5                | 1263             | 166.5            | 1.5                         | 1±1               |
| Concentrated HCl, 12 M   | 70.0                     | 12.0                | 1190             | 167.1            | 0                           | 1±1               |
| Water                    | 72.0                     | 78.4                | 1000             | 168.8            | 0                           | 1±1               |
| Concentrated NaOH, 20 M  | 85.0                     | 55.0                | 1560             | 170.2            | 0                           | 1±1               |
| Concentrated KOH, 20 M   | 98.0                     | 46.8                | 1502             | 171.1            | 0                           | 1±1               |
**Table S2.** Comparison of performances between photopyroelectric and conventional digital microfluidics.

| Techniques                        | Maximum instantaneous speed (mm s\(^{-1}\)) | Maximum continuous moving speed (mm s\(^{-1}\)) | Liquid surface tension (mN m\(^{-1}\)) | Droplet size (µl) | Maximum movable protein (BSA) concentration (mg ml\(^{-1}\)) |
|-----------------------------------|---------------------------------------------|-----------------------------------------------|---------------------------------------|------------------|------------------------------------------------------------|
| Photopyroelectric platform        | 150                                         | 5                                             | 18.9~98.0                             | 0.001~1000       | 20 (movable)                                                |
| Conventional digital microfluidics ([31, 41-43]) | 4.5~40.0                                    | 4.4~36.6                                      | 49.0~72.0                             | 0.4~12.0(restricted by electrode size) | 0.005 (pinning due to biofouling)                           |
Movie S1. Manipulation of silicone oil, \( n \)-hexadecane, \( n \)-decane, \( n \)-heptane, ethanol, and isopropyl alcohol droplets.

Movie S2. Bouncing of a 5 µl \( n \)-heptane droplet on the superomniphobic surface.

Movie S3. Damping oscillation of a 5 µl water droplet.

Movie S4. Continuous manipulation of droplets with diverse volumes.

Movie S5. Merging of two water droplets.

Movie S6. Dispensing of silicone oils and splitting of an ethanol droplet.

Movie S7. Droplets run uphill.

Movie S8. Droplet climbs vertical wall.

Movie S9. Cargo carrier with droplet wheels and driven by light.

Movie S10. Lossless transportation of concentrated BSA droplet.
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