Asymmetric thermal optofluidics based on plasmonic multilayered nanostructures

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ABSTRACT
Manipulating thermo-convective fluid flow induced by plasmonic nanostructures under light illumination has garnered significant attention in various fields, such as biomedical sensing, particle trapping, and drug delivery. However, achieving symmetric optical manipulation of fluid flow encounters challenges in certain applications due to the inherent temporal and spatial symmetry in the energy transfer process. Here, a design of plasmonic nanostructures is proposed to achieve a platform for the asymmetric manipulation of thermally induced fluid flow in an optofluidic environment. The difference in fluid flow rate between forward and backward directions is due to the combined effect of the local asymmetry of the heat transfer in multilayer plasmonic nanostructure and nonreciprocity. The nonreciprocity originates from the violation of time-symmetry due to the temperature gradient-induced convection. We show that the asymmetric convective flow can also be achieved when the size of the plasmonic structure enlarges from nanometer to micrometer, and it can be used for efficient particle separation or transportation in microfluidic systems. Our findings expand the scope of optofluidic applications and stimulate the exploration of design approaches for optical devices.

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Microfluidics, which can precisely manipulate minute volumes of fluid on a microscale, has become a useful tool for biochemical sensing and cell sorting. With the development of optical technology, optofluidics, which integrates photonics and microfluidics, is gradually playing a critical role in developing tunable and integrated fluid control devices. Compared to traditional microfluidics for fluid manipulation, optofluidics is advantageous because it does not require mechanical pumps or physical valves and can be operated contactless and sterile. Furthermore, light excitation offers precision, reconfigurability, and seamless integration with on-chip devices. Researchers have proposed diverse manipulation methods, including the Marangoni effect, thermophoresis, and plasmon-induced photothermal effect, to achieve optofluidic functions like particle and molecule manipulation. Marangoni convection, driven by temperature-dependent surface tension gradients, controls particle aggregation and transportation. The solute concentration gradient at liquid–gas interfaces also affects the surface tension gradient. Thermophoresis utilizes temperature gradients to exert mechanical forces on suspended particles in fluids, driving their migration. However, this process is susceptible to particle size, surface chemistry/charge, and pH value. Alternatively, plasmon-induced thermal convection, driven by the photothermal effect of nanostructures, generates a convection flow that traps particles along streamlines, compelling their movement. Among these optothermal-hydrodynamic strategies, plasmon-induced thermal convection stands out for its straightforward working principle and minimal influencing parameters, offering better controllability.

Optical manipulation platforms utilizing plasmon-induced thermal convection can be categorized into two approaches. The first relies on the local photothermal effect of colloidal plasmonic nanoparticles, where dispersed nanoparticles in a liquid emit thermal energy upon light interaction, creating a uniform temperature gradient for self-driven fluidic systems. However, the uneven distribution of particles leads to an uncontrolled heat source distribution, limiting manipulation of localized thermal energy. Alternatively, artificial nanostructures serve as heat sources to establish desired temperature gradients, typically in lab-on-chip devices immersed in a fluidic environment. Efforts have focused on understanding and manipulating convection origins and motions. For instance, previous studies used single nanodisks to induce fluid convection with varying fluid velocities. However, the fluid velocity in this system is relatively...
slower. An array of bowtie nanoantennas can generate electromagnetic heating to produce thermal convection for higher fluid velocity.\textsuperscript{15} We demonstrated that lattice nanostructures could also manipulate fluid convection.\textsuperscript{21,22} Despite these efforts, current optofluidic platforms can only achieve symmetric properties in fluid control and particle manipulation, highlighting the need to break this symmetry for more flexible flow control and particle motion.

Here, a platform to achieve asymmetric fluid control and distinct particle motions under forward and backward illumination is proposed. The local asymmetry of multilayer plasmonic nanostructures, combined with the nonreciprocity induced by convection flow resulting from differential temperature gradients, enables asymmetrical fluid control. This asymmetry is adjustable by external stimuli applied to the fluid environment, allowing for tuning of fluid viscosity, density, or thermal conductivity. It is a promising microfluidic platform for particle separation or transport. We present a plasmonic-based approach to asymmetrically control thermal convection flow for the next generation of optofluidic-based technologies and applications.

When a nanostructure is excited by a monochromatic wave with its resonant frequency $\omega$, the heat generation stems from the temperature density $q(r)$ inside the nanostructure, where $q(r)$ can be written as\textsuperscript{16}

$$q(r) = \frac{1}{2} \text{Re}[\mathbf{J}(r) \cdot \mathbf{E}(r)] = \frac{\omega}{2} \text{Im}(\varepsilon_0) |\mathbf{E}(r)|^2,$$  

where $\mathbf{J}(r)$ is the local induced current density, which is converted to the heat losses via the Joule heating effect, $\mathbf{E}(r, t) = \text{Re}[\mathbf{E}(r) \cdot e^{i\omega t}]$ is the electric field, $\mathbf{E}(r)$ is its complex amplitude, and $\varepsilon_0$ is the permittivity of the material. The process produces a temperature distribution $T(r, t)$ inside the nanostructure, which is governed by the heat transfer equation,\textsuperscript{16}

$$\rho_m c_m \frac{\partial}{\partial t} T(r, t) - \kappa_m \nabla^2 T(r, t) = q(r),$$  

where $\rho_m$, $c_m$, and $\kappa_m$ are density, specific heat capacity at constant pressure, and thermal conductivity of the plasmonic metal, respectively. $T(r, t)$ is the spatial temperature distribution inside the plasmonic nanostructure. Since the absorption of light by the surrounding fluid is almost zero, it can be ignored, i.e., $q(r) \approx 0$. However, other modes of heat transfer, such as convection, may still be present. Therefore, the temperature distribution in the surrounding fluid is

$$\rho_c c_s \left[ \frac{\partial}{\partial t} T(r, t) + \nabla \cdot \left( \mathbf{v}(r, t) \nabla T(r, t) \right) \right] - \kappa_c \nabla^2 T(r, t) = 0,$$

where $\mathbf{v}(r, t)$ is the fluid velocity and $\rho_c$, $c_s$, and $\kappa_c$ are mass density, specific heat capacity at constant pressure, and thermal conductivity of the surrounding fluid, respectively.

An increase in the temperature of the fluid around the nanostructure will reduce its density, creating upward fluid convection, which is governed by the Navier-Stokes equation,\textsuperscript{16}

$$\frac{\partial}{\partial t} \mathbf{v}(r, t) + \mathbf{v}(r, t) \cdot \nabla \mathbf{v}(r, t) = -\nabla p + \nu \nabla^2 \mathbf{v}(r, t) + \mathbf{f}_f(T(r, t)),$$

with continuity equation $\nabla \cdot \mathbf{v} = 0$, where $\nu$ is the kinematic viscosity of the fluid and $\mathbf{f}_f$ is the volume force per unit mass caused by the temperature gradient. The Boussinesq approximation is applied to reduce computing complexity,\textsuperscript{16}

$$\mathbf{f}_f(T) = \beta_p (T(t, t) - T_0) \hat{\mathbf{z}},$$

where $\beta_p$ and $T_0$ are the dilatation coefficient of fluid and initial temperature, respectively, and $g$ and $\hat{\mathbf{z}}$ are the gravitational acceleration and the upward $z$-direction unit vector, respectively.

Based on the aforementioned equations, one can get plasmonic thermal convection flow. Next, the structural design concept for realizing asymmetric fluid flow is discussed. As shown in Fig. 1(a), we propose a system comprising a plasmonic nanostructure. The nanostructure consists of stacked layers of metallic asymmetric nanodisks separated by a SiO$_2$ spacer layer, eventually forming two stacked layers on the left and right sides. The whole nanostructure immersed in a fluid is mounted on a SiO$_2$ glass. When a single layer of asymmetric nanodisks is excited by its two resonant frequencies, different nanodisks can act as heat sources, inducing wavelength-selective thermal convection. However, if a single layer is illuminated by the same plane wave from two opposite propagation directions, the same nanodisk is excited as a heat source, resulting in identical convection. To overcome this limitation, we propose stacking a monolayer with two elements into a multilayer to create two heat transfer channels (left- and right-stacked layers). This configuration allows the same incident light beam to interact with the two channels. Under uniform illumination from the forward and backward directions, the left- and right-stacked layers will exhibit different thermal dissipation, leading to a temperature difference with opposite symbols in the horizontal direction and generating different convections [Fig. 1(b)]

Our design principle is evident in the electric field and power dissipation distribution within the nanostructures [Figs. 1(c) and 1(d)]. When illuminated with the resonant wavelength of the small nanodisk, asymmetric light absorption occurs, mainly in the top nanodisk of the left-stacked layers for forward incidence and in the bottom nanodisk of the right-stacked layers for backward incidence. This results in nonuniform absorbed energy across the nanostructure. Calculations are performed using COMSOL 6.0, with additional simulation details provided in the supplementary material S1. Feasibility analysis is presented in the supplementary material S2.

We present the asymmetrical manipulation of thermal convective flow in a 50% glycerol-water solution (refractive index $n_2 = 1.4$, nearly the same as the substrate’s refractive index). The absorption cross section of the system is shown in the supplementary material S3. Based on the multiphysics coupling simulation, convective flow patterns overlaid on temperature distributions are calculated in Figs. 2(a) and 2(b). Under forward illumination, resonance occurs on the top of the left-stacked layers, where the heat is mainly produced and subsequently transferred to the left side of the nanostructure. In contrast, the resonance occurs on the bottom of the right-stacked layers under backward illumination, where the heat is generated and transferred to the right side of the nanostructure. The reversed temperature gradient in surroundings and asymmetric shape streamlines along the horizontal direction between forward and backward directions can, thus, be obtained. When the light is excited in the forward direction, it initially interacts with the top nanodisk on the left-stacked layers next to the fluid. For backward, it first interacts with the bottom nanodisk on the right-stacked layers next to the substrate. The thermal conductivity of the fluid and substrate is different, resulting in different heat
generation and conduction in the two cases. Therefore, one of the contributions to the difference in fluid flow is the asymmetry of the heat transfer in multilayer plasmonic nanostructure.

In Fig. 2(a), the direction of thermal energy transfer is opposite to that of the fluid flow. In contrast, in Fig. 2(b), the direction of the thermal energy transport through the nanodisks is along the direction of the fluid flow. The clockwise/counterclockwise motion convective flow provides a tangential momentum that can be interpreted as a time-odd bias to the energy or fluid flow in the vertical direction along the z-direction. If one imagines two points placed along the flow, it is clear that the speed of the fluid along the flow differs from that against the flow. Thus, the transmitted energy along or against the flow differs. Therefore, another contribution to the difference in fluid control is the nonreciprocity. This difference in temperature and velocity increases with the biasing element, which is the incident laser intensity (supplementary material S4). Figures 2(c) and 2(d) plot the asymmetric temperature and velocity as a function of illumination time. The system does not show an instantaneous difference in temperature and velocity but gradually increases with time. Moreover, the heat transfer rate is always faster from front to back than from back to front of the plasmonic structures. Relatively higher temperature and velocity values are acquired in the forward direction than in the backward direction with the substrate. This difference in temperature and velocity reaches an asymptotic value when the system approaches a steady state. The asymmetric optofluidic results could also obtained by illuminating with the resonant wavelength of the big nanodisk (supplementary material S5).

Figures 3(a)–3(c) depict the spatial temperature distributions of the nanostructure. When the excitation light is incident from the forward, the left-stacked layers attain a higher temperature than the right-stacked ones. However, the right-stacked layers attain a higher temperature for the inverse process than the left-stacked layer. This performance of separation of heat energy between the two branches of the nanostructure is very significant and efficient in affecting the fluid flow rate to attain an asymmetric velocity. Vertical and horizontal 2D slices of the flow velocity field distributions are depicted in Fig. 3(d). We see a higher flow velocity distributed around the left half of the chamber under the forward illumination case. For the reversed illumination case, the right half shows higher flow velocity. Importantly, these flow velocity field distributions will act on nanoparticles suspended in the fluid and affect their trajectories. These unique performances in temperature and velocity can be attributed to the occurrence of asymmetry, which is also evidenced in line plots along the y [Fig. 3(b)] and z [Fig. 3(c)] axes and 2D color maps of y–z [Fig. 3(d)] and x–y [Fig. 3(f)] cut planes. In addition to the steady-state results, we also provide the transient temperature of the system as a spatial distribution (see supplementary material S6).
Next, we explore the factors that influence the disparity in temperature and velocity. Figure 4 depicts the temperature and velocity difference between forward and backward illumination as a function of viscosity change [Fig. 4(a)], heat capacity change [Fig. 4(b)], density change [Fig. 4(c)], and thermal conductivity change [Fig. 4(d)]. The variation amplitude of these parameters is 5%, 10%, 15%, 20%, and 25% based on the original parameters (see supplementary material S7). External stimuli, such as an external magnetic field, can achieve these modifications. The results reveal that the thermal conductivity of fluid has the greatest influence on temperature asymmetry. When the change in magnitude of the fluid’s thermal conductivity increases, the temperature difference increases; correspondingly, a strong change in the velocity difference will be obtained. The fluid’s viscosity and density significantly affect the velocity difference. Specifically, the increase in the amplitude of the viscosity change will lead to an increase in the velocity difference. In contrast, increasing the density-gradient amplitude will decrease the velocity difference. The fluid’s viscosity, density, and thermal conductivity should be optimized to enhance difference and, thus, benefit asymmetrical fluid control applications.

Considering that the velocity of the fluid induced by the present nanostructure is too small to induce any molecular motion, we explore the possible applications by changing the present structure into a microsized structure in the following part (the simulation details are in the supplementary material S1). The asymmetric convective flow can be used to manipulate the nanoparticles. The motion of particles originates from the drag force, buoyancy, and gravitational forces (the force analysis and equations are shown in the supplementary material S8). We calculate the trajectories of SiO$_2$ particles in a microfluidic chamber, as shown in Fig. 5(a) (the asymmetrical fluid velocity is given in the supplementary material S9). The particles released in the center position of the system take different trajectories under different direction illuminations (supplementary material Video 1 and supplementary material Video 2). Under forward and backward illumination, particles driven by balanced force are transported in circles from the initial positions to the right and left sides of the microfluidic chamber following the streamline, respectively. The opposite particle trajectory in different light directions suggests that our asymmetrical optofluidic device can transport and separate particles.

We then investigate factors that affect particle speed. Figures 5(b)–5(d) show the particle speed as the function of particle size, inter-particle distance, and particle type. The particle velocities change slightly when the particle size changes from 60 to 160 nm [Fig. 5(b)]. In addition, the effect of the inter-particle distance on the particle speed also exhibits very small changes [Fig. 5(c)]. Meanwhile, the study of three different types of particles [polystyrene (PS) ($\rho_{PS} = 1.06$ g/cm$^3$), SiO$_2$ ($\rho_{SiO_2} = 2.2$ g/cm$^3$) and ZrO$_2$ particles ($\rho_{ZrO_2} = 5.85$ g/cm$^3$)] shows that the density of particles has little effect on the particle speed [Fig. 5(d)]. Figure 5(e) plots the particle speed of different input power. As the input power increases from 6 to 16 mW, both forward and backward velocities of the particles increase, and the ratio of the two remains roughly stable.

Furthermore, we explore the influence of the system on particle velocity [Figs. 5(f) and 5(g)]. Figure 5(f) illustrates the relationship between particle speed and liquid level height. Both forward and backward particle speeds increase gradually with the rise in liquid level height. Figure 5(g) demonstrates the impact of different nanodisk gaps.
FIG. 3. Temperature distributions of nanostructure along (a) x direction \((y = 0 \text{ nm, } z = 165 \text{ nm})\), (b) y direction \((x = 0 \text{ nm, } z = 165 \text{ nm})\), and (c) z-direction \((x = 0 \text{ nm, } y = 0 \text{ nm})\). Velocity field distributions of convection in (d) x-z cut plane \((y = 0 \text{ nm})\), (e) y-z cut plane \((x = 0 \text{ nm})\), and (c) x-y cut plane \((z = 165 \text{ nm})\). In (a), the blue and pink boxes represent the positions of the left-stacked and right-stacked layers. In (a)–(c), the inset is a schematic of the structure with coordinate axes. In (d)–(f), white arrows illustrate the directions of convection. The plane where \(z = 165 \text{ nm}\) is the upper surface of the nanostructures. All data are given at a steady state.

FIG. 4. Temperature and velocity difference between forward and backward illumination as a function of (a) viscosity change, (b) heat capacity change, (c) density change, and (d) thermal conductivity change. All the temperature and velocity differences are given at steady state.
on particle speed. As the gap increases, forward and backward particle speeds initially rise sharply before slowing down. Concurrently, the ratio decreases progressively, suggesting that a larger gap induces relatively less asymmetry. In summary, particle velocity is minimally affected by particle properties, but greatly affected by incident light power and system characteristics. Future work can explore tunable gradient index optics with stacked asymmetric nanodisks to manipulate light beams and influence fluid convection patterns. Photothermal modulation techniques can also dynamically adjust fluid temperature distributions for controllable convection. Another avenue is to investigate bioinspired design strategies, potentially leading to microscale actuators for precise fluid motion control in response to light stimuli. These approaches hold promise for advancing fluid dynamics control and merit further exploration.

In conclusion, we demonstrated an asymmetrical optofluidic platform based on a plasmonic multilayer nanostructure. The asymmetry in our system manifests in the fluid velocity and temperature of thermal convection, favoring the forward direction over the backward direction. The physical mechanism is due to the multilayer nanostructure’s local asymmetry and the nonreciprocity provided by convection flow caused by different temperature gradients. Moreover, strong asymmetry can be obtained by increasing the incident optical intensity or applying external stimuli for a fluid to optimize the fluid characteristics. Calculations indicate that as the size of the plasmonic nanostructure in our platform increases from nanometers to micrometers, it becomes capable of particle manipulation for particle separation or transportation. We envision that as microfluidic integration improves, this system will unlock opportunities for optofluidic applications and inspire the exploration of innovative approaches for optical devices.

FIG. 5. (a) SiO₂ particle trajectories under forward and backward illumination cases. All particles have a uniform diameter of 100 nm and a uniform spacing of 50 nm. The average particle speed as a function of (b) particle size, (c) interparticle distance, (d) particle type, (e) input power, (f) liquid height, and (g) disk gap. All particle speeds are the average values of the maximum particle speeds.
schematic of force analysis of particles in fluid convection, and simulated fluid velocity in particle trajectory simulation. See supplementary video 1 and supplementary video 2 for different particle trajectories under different direction illuminations.

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AUTHOR DECLARATIONS
Conflict of Interest
The authors have no conflicts to disclose.

Author Contributions
Zhimin Jing: Data curation (lead); Formal analysis (lead); Investigation (lead); Writing – original draft (lead). Cuiping Ma: Investigation (supporting); Writing – review & editing (supporting). Peihang Li: Investigation (supporting); Visualization (supporting). Peng Yu: Writing – review & editing (supporting). Arup Neogi: Conceptualization (lead); Writing – original draft (supporting); Writing – review & editing (equal). Zhiming Wang: Conceptualization (equal); Supervision (lead); Writing – review & editing (equal).

DATA AVAILABILITY
The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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