

## Regular Article

# Transport and trapping of nanosheets via hydrodynamic forces and curvature-induced capillary quadrupolar interactions



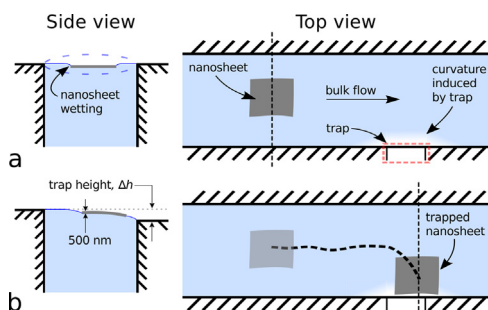
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## GRAPHICAL ABSTRACT



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## ABSTRACT

**Hypothesis:** The manipulation of nanosheets on a fluid-fluid interface remains a significant challenge. At this interface, hydrodynamic forces can be used for long-range transport ( $>1 \times$  capillary length) but are difficult to utilize for accurate and repeatable positioning. While capillary multipole interactions have been used for particle trapping, how these interactions manifest on large but thin objects, i.e., nanosheets, remains an open question. Hence, we posit hydrodynamic forces in conjunction with capillary multipole interactions can be used for nanosheet transport and trapping.

**Experiments:** We designed and characterized a fluidic device for transporting and trapping nanosheets on the water-air interface. Analytical models were compared against optical measurements of the nanosheet behavior to investigate capillary multipole interactions. Energy-based modeling and dimensional analysis were used to study trapping stability.

**Findings:** Hydrodynamic forces and capillary interactions successfully transported and trapped nanosheets at a designated trapping location with a repeatability of 10% of the nanosheet's length and 12% of its width (length = 1500  $\mu\text{m}$ , width = 1000  $\mu\text{m}$ ) and an accuracy of 20% of their length and width. Additionally, this is the first report that surface tension forces acting upon nanoscale-thick objects manifest as capillary quadrupolar interactions and can be used for precision manipulation of nanosheets.

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## 1. Introduction

Two-dimensional nanomaterials, also called “nanosheets”, feature thicknesses on the order of nanometers, with lengths and widths on the order of micrometers to millimeters. The fabrication

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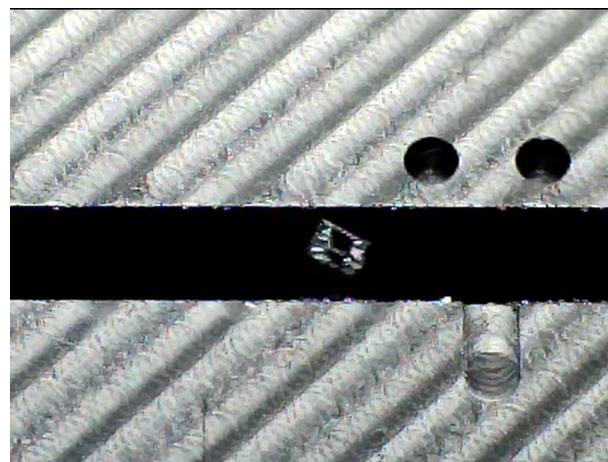
and manipulation of nanosheets, as well as their integration with substrates and devices, remains a significant challenge, despite its relevance to a broad range of fields, from nanotechnology, to neuroscience, to materials science [1–10]. Nanosheets hold promise to bring significant advances in thin-film transistor, actuator, energy harvesting/power generating, and opto-electronic technologies [1–5]. In recent years, methods of nanosheet fabrication involving self-assembly of nanomaterials at liquid-air interfaces have been shown to be reliable for nanosheet manufacturing [7–10]. However, objects on liquid-air interfaces are subject to a complex combination of capillary and hydrodynamic forces [11–22].

Thus, while the ability to fabricate nanosheets has been well established, the manipulation of nanosheets remains an unsolved problem. Within a single fluid, there are many options for particle manipulation, ranging from optical, magnetic, electrokinetic, closed-loop microfluidic hydrodynamic, and acoustic systems [23–30]. Yet, at fluid-fluid interfaces, options are largely limited to hydrodynamic and capillary forces. Curvature-induced capillary multipole interactions at the fluid-fluid interface are able to manipulate a variety of particles but typically across distances less than one capillary length [11–22]. From prior literature, the dominant multipole interaction that facilitates particle movement varies depending on the geometry of the object: quadrupolar capillary interactions are known to facilitate the attraction of small ( $r \sim 1 \mu\text{m}$ ) particles, while monopolar capillary interactions are known to facilitate the attraction of large ( $r \sim 1 \text{mm}$ ) isometric objects [11–22]. For extremely anisotropic materials with aspect ratios much greater than one—e.g., nanosheets—it remains unclear which multipole capillary interaction is the dominant term. Nonetheless, for distances greater than one capillary length, multipole capillary interactions are generally impractical for rapid manipulation of nanosheets. Conversely, hydrodynamic forces are effective for long-range transportation at the air-water interface but are difficult to utilize for trapping.

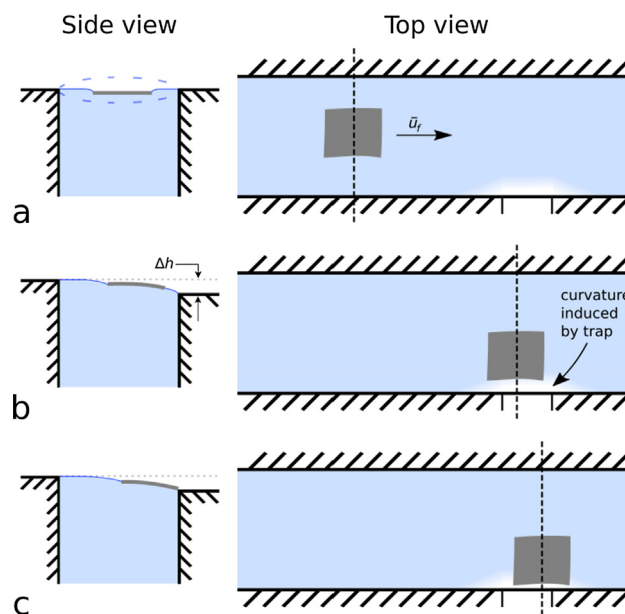
Ultimately, we present an approach that combines hydrodynamic and curvature-induced capillary interactions in a device for the transport and trapping of nanosheets at a liquid-air interface. In our device, the hydrodynamic forces, produced by bulk fluid motion, enable long-range ( $>1 \times$  capillary length) transport while the capillary interactions, produced by a curvature-inducing trapping feature, enable short-range ( $<1 \times$  capillary length), accurate and repeatable trapping thereby passively manipulating nanosheets on the liquid-air interface. Additionally, we demonstrate, with modeling and experiment, the interactions of nanosheets via quadrupolar capillary interactions.

## 2. Materials and methods

A schematic of our device is shown in Fig. 1. Initially, a nanosheet is placed onto a flat water-air interface, thus deforming the water surface around itself. Due to surface tension forces, the nanosheet remains trapped at the water-air interface. Bulk flow is produced within the channel, and the nanosheet is transported “downstream” via hydrodynamic forces, as indicated in Fig. 1a, right. Further downstream, a trap is created from a micro-machined notch in the channel wall, which induces curvature on the water-air interface, shown in Fig. 1b. As the nanosheet approaches the trap, the curvature induced by the nanosheet interacts with the curvature induced by the trap, attracting the nanosheet towards the trap. Ultimately, the nanosheet comes to rest at the trap, as shown in Fig. 1c. For further illustration, a video of nanosheet manipulation is shown in Supplemental Movie 1.



Movie 1.



**Fig. 1.** The nanosheet trapping device comprises a water-filled, open millifluidic channel with a notch (i.e., trap) along one edge. As a nanosheet flows through the channel, it comes to rest in contact with the trap. The device works by using a combination of hydrodynamic forces far from the trap (view a) and curvature-induced capillary interactions close to the trap (views b, c) to transport and trap nanosheets, respectively. Top views illustrate the position of the nanosheet in the channel relative to the trap. Side views, at locations indicated by vertical dashed lines, illustrate curvature of water-air interface. (a, side view) The trap water height is set properly when the fluid level has minimal curvature. (a, top view) The water and nanosheet both flow with average velocity,  $\bar{u}_r$  far from the trap (approximately  $1\text{--}10 \times$  capillary length of the water-air interface, or equivalently  $2.7\text{--}27 \text{mm}$ ). (b, side view) Near the trap ( $<1 \times$  capillary length ( $<2.7 \text{mm}$ ) of the water-air interface), the nanosheet's trajectory is influenced by water-air curvature arising from the trap's height difference,  $\Delta h$ . (c, side view) The nanosheet comes to rest at the trap where the water-air interface surface energy is minimized.

### 2.1. Device design and fabrication

We designed and fabricated a device for nanosheet transport and trapping, shown in Fig. 2. The device was designed in computer aided design software (SolidWORKS); G-code was generated using computer-aided manufacturing software (HSMWorks). The device was fabricated using a CNC mill (Haas Office Mill, Model

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