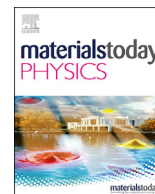




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Marangoni convection-driven laser fountains on free surfaces of liquids

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ABSTRACT

It is well known that an outward Marangoni convection from a low surface tension region will make the free surface of a liquid depressed. Here, we report that this established perception is only valid for thin liquid films. Using surface laser heating, we show that in deep liquids a laser beam pulls up the fluid above the free surface generating fountains with different shapes, and with decreasing liquid depth a transition from fountain to indentation with fountain-in-indentation is observed. High-speed imaging captures a transient surface depression before steady elevation is formed, and computational fluid dynamics simulations reveal the underlying flow patterns and quantify the depth-dependent and time-resolved surface deformations. Systematic investigation of the effect of laser parameters, surface tension and area of the fluid on its surface deformation further confirms that the laser fountain is a result of dynamic competition between outgoing Marangoni convection and the upward recirculation flow. Experiments and simulations also reveal that a smaller surface area can dramatically strengthen laser fountain. The discovery of laser fountain and the development of related experimental and simulation techniques have upended a century-old perception and opened up a new regime of interdisciplinary research and applications of Marangoni-induced interface phenomena.

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1. Introduction

The Marangoni effect is the convection of fluid at a free liquid surface driven by a surface tension gradient. Marangoni-induced flow patterns and surface deformation have been attracting enormous attention due to their fascinating complexity and enormous applications in dynamic fluid control [1–4]. Since the Marangoni convection pulls liquid away from the lower surface tension region, it has been observed and widely accepted that the surface with a

locally low tension will become depressed [1–6]. Tears of wine and Bénard cells, induced by composition and temperature gradient, respectively [5], are prominent examples of Marangoni convection-driven surface depression although it took 50 years to correctly set apart the crucial role of surface tension in Bénard cells from contributions due to natural convection [7,8]. The first quantitative description of surface depression was provided by Landau by considering the lateral flow only for a given surface tension gradient in a thin liquid film [6]. More rigorous theoretical treatments [9–16] and controlled experiments [10,17–21] were performed later with non-uniform surface tension created by either substrate heating or surface radiative heating. Besides these fundamental investigations, more cases have been investigated in applications involving liquids or soft matters such as lithography and 3D printing [22–24], heat transfer and mass transport [19], crystal growth and alloy welding [25,26], dynamic grating and spatial light modulator [15,27], and microfluidics and adaptive optics [19,28–30]. Although these studies provided a detailed description of surface depression and flow patterns, they have failed to go beyond Landau's theory and offer a unified theory of surface deformation in liquid with an arbitrary depth.

2. Results and discussion

In this work we use a relatively low-power (<1 W in general) continuous-wave (CW) laser beam to create a non-uniform surface temperature field to induce the Marangoni effect. Compared with contact heating from the wall substrates, surface laser heating is noncontact and has the advantage of precise control of temperature in space and time without inducing natural convection, which is especially useful in a deep liquid. Fig. 1A shows a schematic of our experimental setup. Ferrofluid is held in a petri dish and is illuminated from above with a laser beam. Unless otherwise stated, the laser's wavelength is 532 nm, and a 532-nm notch filter is used to block the laser for better imaging. The resulting distortions of the fluid surface are imaged with digital cameras. Commercial ferrofluid (Educational ferrofluid EFH1 from Ferrotec Corporation, see *Supplementary data* for detail) is chosen because of its high light-absorbing property for efficient surface heating. The laser beam is either unfocused, or is slightly focused with a cylindrical lens or an axicon lens to create a line beam and a ring beam, respectively.

Ferrofluid is a so-called “magic” liquid and is best known for its astonishing surface spikes generated by a magnet [31]. Surprisingly, similar spikes can also be created by a laser. Fig. 1B and C show two spikes or cones on the flat surface of ferrofluid under unfocused laser beam. Like in a magnetic field, the height of the liquid cone can be adjusted by tuning the laser power, and the cone is stable as long as the laser stays on. What is different is that other surface protrusions can be created by simply varying the laser beam shape. Fig. 1D–H show examples of a ridge and a hat created by a line beam and a ring beam, respectively. A taller cone can be created by a tightly focused ring beam (Fig. 1I). Because their shapes can be controlled by the shape of the laser beams, we call these surface protrusions “laser fountains”. Like a mechanically pumped fountain, once the laser (the virtual pump) is turned off, the fountain will vanish in less than 0.1 s (*Supplementary Video S1*). It should be noticed that when the liquid layer is shallow, the same laser beams actually create indentations (Fig. 1J–N) even for the same ferrofluid [2,3,10,14]. A higher laser power or a shallower layer will make depression so strong that the entire liquid layer become ruptured (Fig. 1K, M and N) [2,3,10,14]. We note that the extent of deformation demonstrated here is qualitatively different than the surface depressions previously reported in the literature, which were too weak to be directly observed by naked eyes or ordinary photography [10,17–21].

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To understand these distinct deformations between deep and shallow liquids, we varied the liquid layer thickness while keeping the laser beam the same. Fig. 2A–D show that an indentation is observed when the thickness of ferrofluid is 2.8 mm, in agreement with the observation made for a shallower ferrofluid layer in Fig. 1J–N as well as with the findings in past studies [2,3,10,14]; however, in a 3.2-mm thick liquid, a cone begins to emerge from the center of the indentation. With increasing thickness, the cone continues to grow and the indentation becomes less and less visible. When the liquid thickness reaches 8 mm or above, the cone dominates the surface and the indentation disappears completely, i.e., a laser fountain is formed. The laser fountains and the depth-dependent transition from surface indentation to laser fountain have never been reported in the literature even in deep liquids, probably because they are not anticipated by any existing theory.

Despite these seemingly stationary surface deformations, the fluid underneath actually is experiencing a rapid circulation. To confirm this, we added fluorescent tracer microspheres in the liquid to probe the Marangoni convection [32], and, indeed, we observed the same outward flow for both surface depression (*Supplementary Video S2*) and surface elevation (*Supplementary Video S1*). Based on the video of tracers in *Supplementary Video S1*, the outward liquid has a high speed of more than 2 cm/s [32], making the cone a true fountain instead of simple surface deformation. Clearly, these divergent flows are Marangoni convection induced by local laser heating. To understand why similar divergent Marangoni flow leads to totally different surface deformation, we employed a high-speed camera to capture the surface profiles right after the laser illuminates a deep ferrofluid. The time-resolved images shown in Fig. 2E–H clearly reveal the draining of fluid and the formation of a depression due to the Marangoni convection before the appearance and slow growth of a cone. Thus, there is no simple relationship between the outward Marangoni flow and surface depression as previously understood.

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We emphasize that there have been numerous attempts to understand the Marangoni flow-driven surface deformation, but no existing theory can predict the deformation patterns of a liquid with an arbitrary depth in a straightforward manner. Various approximations often have to be made even in analyzing surface deformation of a shallow liquid, due to complexities in solving the highly nonlinear Navier-Stokes equations, especially when the flow is driven by surface tension gradient and coupled with heat transfer and optical absorption. To avoid the limitations of oversimplified analytical models, we turned to computational fluid dynamics (CFD), which allows us to solve the complete optical-thermal-flow coupled problem with a free boundary. Here we modeled the laser to be coupled with ferrofluid through its optical absorption. Note that although ferrofluid is a composite nanofluid made of superparamagnetic Fe_3O_4 nanoparticles suspended in kerosene, it does not exhibit any magnetism in the absence of an external magnetic field and can still be well modeled as a homogeneous single-phase liquid [31]. Fig. 3A shows the COMSOL model of a ferrofluid layer, which assumes a cylindrical symmetry under a vertically incident Gaussian laser beam. Details of the COMSOL model and parameters are included in the *Supplementary data*. The CFD simulation results in Fig. 3B show the expected Marangoni convection and surface indentation in a representative 1.3-mm-thick liquid, the simulation also confirms that the surface depression increases with decreasing thickness until the rupture of the liquid layer – a manifestation of Marangoni instability [2,3,10,14].

Inspired by the successful simulation of surface depression in a

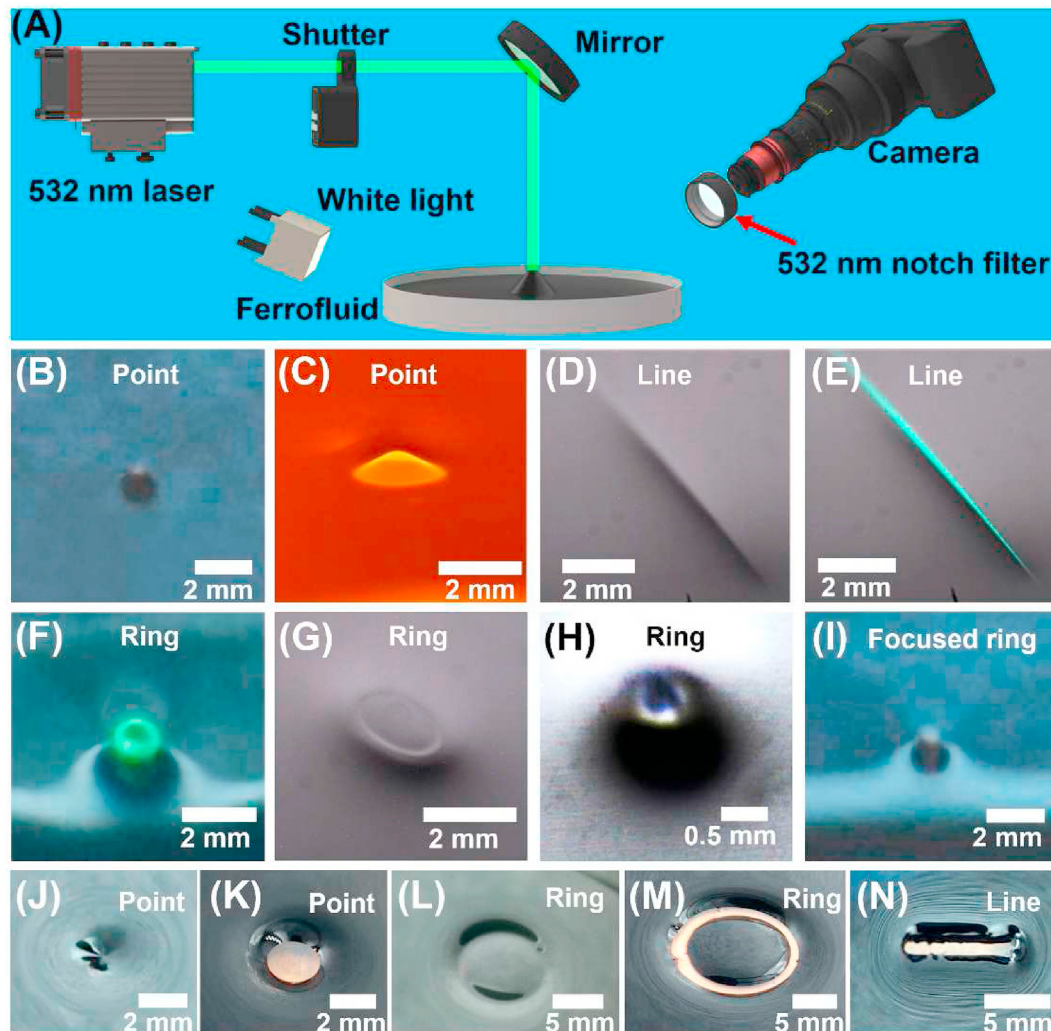


Fig. 1. Experimental setup and photographs of steady-state surface deformations in a ferrofluid generated by 532-nm continuous-wave laser beams. (A) Schematic of experimental setup. (B–H) Fountains with the shape of (B–C) a cone, (D–E) a ridge and (F–H) a hat generated by an unfocused beam, a line-focused beam and a ring beam, respectively. The cone in (I) is generated by a highly focused ring beam. The depth of ferrofluid is 8 mm. (J–N) Surface indentations induced by the same shapes of laser beams in a shallow ferrofluid (0.5 mm). The laser is blocked in all except (C), (E) and (F) by a notch filter. A 532-nm long pass filter is used for (C). The laser beam diameter is 0.6 mm for (B) and 1 mm for (C).

shallow liquid, we increase the depth of ferrofluid in the subsequent simulation. Surprisingly, Fig. 3C–H show that the effect of liquid depth on the surface deformation can also be well verified. A close comparison of flow patterns in the shallow and deep liquids reveals that the surface deformation is not solely induced by the outward Marangoni convection, but, rather, determined by the competition between the Marangoni flow on the free surface and the upward recirculation flow arising from the subsurface colder region due to mass conservation. In the shallow liquid in Fig. 3B, the recirculation flow is hindered by the viscous shear force at the substrate, so a depression is formed. As the liquid depth increases, there will be less flow resistance from the substrate, thus leading to a weaker depression. For a sufficiently deep liquid, the upward recirculation flow may become strong enough to outweigh the outward Marangoni flow, causing a fountain to form.

Understanding the distinct surface deformation in liquids with different depths helps us to unravel the dynamics of the surface deformation process: the observed transient deformation must originate from a dynamic competition between the Marangoni convection and the recirculation flow. This understanding is confirmed by the CFD simulations. Fig. 4A and B show that, initially,

a hot spot exists near the liquid surface that result in the outward Marangoni convection from the laser beam center. Since the capillary flow is highly localized, there is only a very weak recirculation flow and the surface becomes depressed as observed in a shallow liquid. However, as shown in Fig. 4C and D, the recirculation flow develops very quickly and the resulting convection heat transfer causes the surface temperature gradient to decrease. Eventually, the liquid mass carried by recirculation exceeds that by the outward Marangoni flow, leading to the formation of a cone in the center of indentation and ultimately, stationary cones above the flat surface.

It should be noted that the observed laser fountains phenomenon is driven by the Marangoni effect although an upward flow can also be generated by natural convection when a column of liquid is heated volumetrically by a laser beam [33]. The former conclusion is not difficult to reach from the observations presented above. First, because of a high optical absorption coefficient of ferrofluid (867 cm^{-1} at 532 nm), the thickness of the laser heating layer is only about $15 \text{ }\mu\text{m}$, much smaller than the observed elevation heights in Figs. 1 and 2, so the laser heated elevated liquid column hinders rather than induces the natural convection. Second, both

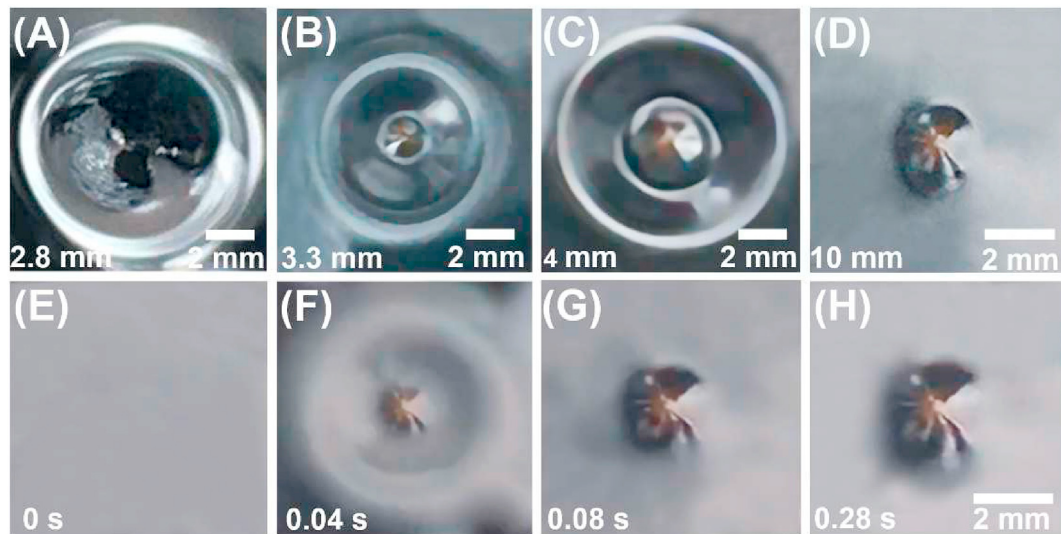


Fig. 2. Images of depth-dependent and time-resolved surface deformations. (A–D) Emergence of a cone fountain from the center of a larger indentation with increasing depth of ferrofluid from 2.8 mm in (A) to 10 mm in (D). (E–H) Time-resolved surface deformation in a deep ferrofluid (8 mm) after the turn-on of the laser showing initial formation of indentation followed by the growth and dominance of a cone. The power of laser is 1 W.

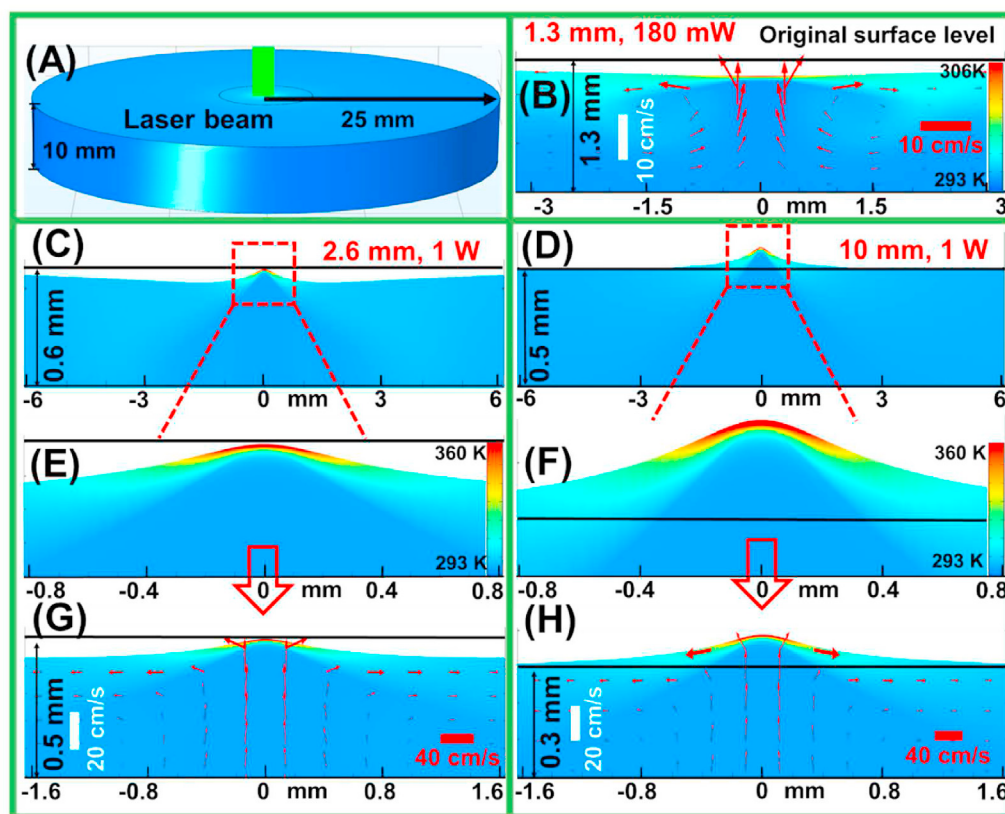


Fig. 3. Stationary surface deformations, temperature distributions and flow patterns from COMSOL simulations with a 0.3-mm diameter laser beam. (A) Simulation configuration with a 10-mm thick liquid. The diameter of liquid domain is fixed at 5 cm. (B–H) Simulated surface deformation, flow and temperature fields in ferrofluid with depth of (B) 1.3 mm, (C, E and G) 3.2 mm and (D, F and H) 10 mm. (C–H) show selected areas for better viewing.

the experiments and simulations have revealed that the surface depression happens before the surface elevation, in other words, the surface Marangoni surface flow is the driving force for the upward flow and fountain in contrast to an initial elevation due to convection. Third, the thickness of ferrofluid is already much larger than the laser penetration depth from the very beginning

(Fig. 2A–D), as the thickness of ferrofluid further increases, the relative thickness of heating layer becomes more and more insignificant. Thus, there is no reason to attribute the surface elevation to the natural laser-induced convection in a deep liquid if we believe that the Marangoni effect is responsible for the surface depression in a shallower ferrofluid layer.

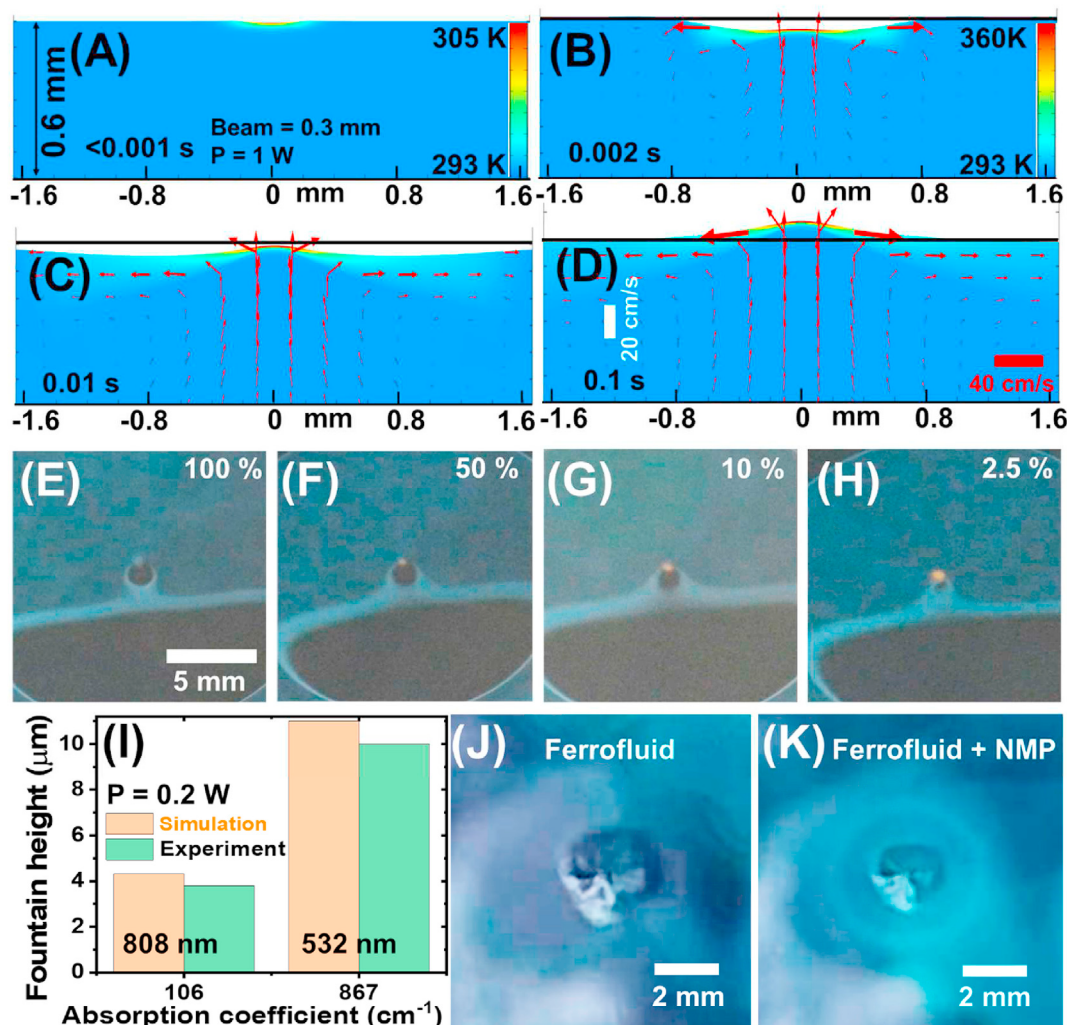


Fig. 4. Simulated transient surface deformation, elevation height with varying absorption coefficient and surface tension. (A–D) Temporally resolved simulated surface profile, temperature distribution and flow velocity in a 10-mm deep ferrofluid. (E–H) Laser fountains in diluted ferrofluids. The percentages are volume concentration of original ferrofluid in kerosene. (I) Fountain heights by 532-nm and 808-nm lasers with 200 mW power and 1 mm diameter. (J–K) Comparison of fountains in (J) ferrofluid and (K) ferrofluid covered by a thin layer of NMP.

To further prove that the laser fountains are driven by surface tension Marangoni flow instead of natural convection, we performed a series of experiments and simulations by reducing surface tension and increasing optical heating depth to investigate how they will affect the height of laser induced fountain. If convection were the main contributor to the phenomenon, the latter modification should result in an increased fountain/elevation height. Fig. 4E–H show a diminished laser fountain under the same 532-nm laser excitation when the ferrofluid is diluted by kerosene. Note that the original ferrofluid is made of ~10% volume percentage of Fe₃O₄ nanoparticles in kerosene. Diluted ferrofluid will have a larger laser penetration depth to heat up the liquid and create more natural convection. Another way to increase laser penetration depth is to increase laser wavelength. Fig. 4I shows that the height of fountain from both experiments and simulations decreases significantly when the wavelength of laser is changed from 532 nm to 808 nm. Here a home-built scanning probe station is used to measure the surface deformation (Fig. S2), and the surface profile of a typical fountain is shown in Fig. S3. In the remaining discussions, only height is used to characterize surface deformation. Finally, we added a thin layer of n-methyl-2-pyrrolidone (NMP) on ferrofluid. NMP is immiscible with ferrofluid and has a lower density, while its

temperature coefficient of surface tension is only about 1/3 that of ferrofluid. Fig. 4J and K show that the size of laser fountain is reduced significantly when the ferrofluid is covered by a thin layer of NMP. The observation of shortened laser fountains when the surface tension is reduced proves the Marangoni effect is the sole driving force for laser fountain. Increasing convection via an increased thickness of laser heated liquid layer did not increase the fountain height and shows a contrasting effect on how natural convection would contribute to the fountain phenomenon.

While it is almost impossible to reduce or eliminate the intrinsic temperature coefficient of surface tension of a liquid in experiment, it is very easy to set it to zero in simulation. Fig. S6 shows that the surface temperature quickly increases to the flash point of ferrofluid in less than 0.01 s if its temperature coefficient of surface tension is reduced to near zero. In reality, using an infrared thermal camera (SC7000, FLIR) [34], Fig. 5A, Fig. S7 and Supplementary video S3 reveal that the surface temperature near the laser point quickly stabilizes due to rapid Marangoni flow and increases only by 12°, in a good agreement with the simulation in Fig. 5B and Fig. S7. Note that the “apparent” low temperature region within a 1 mm radius from the center in Fig. 5A is an artifact and is due to sloped surface of the laser-induced fountain. This temperature

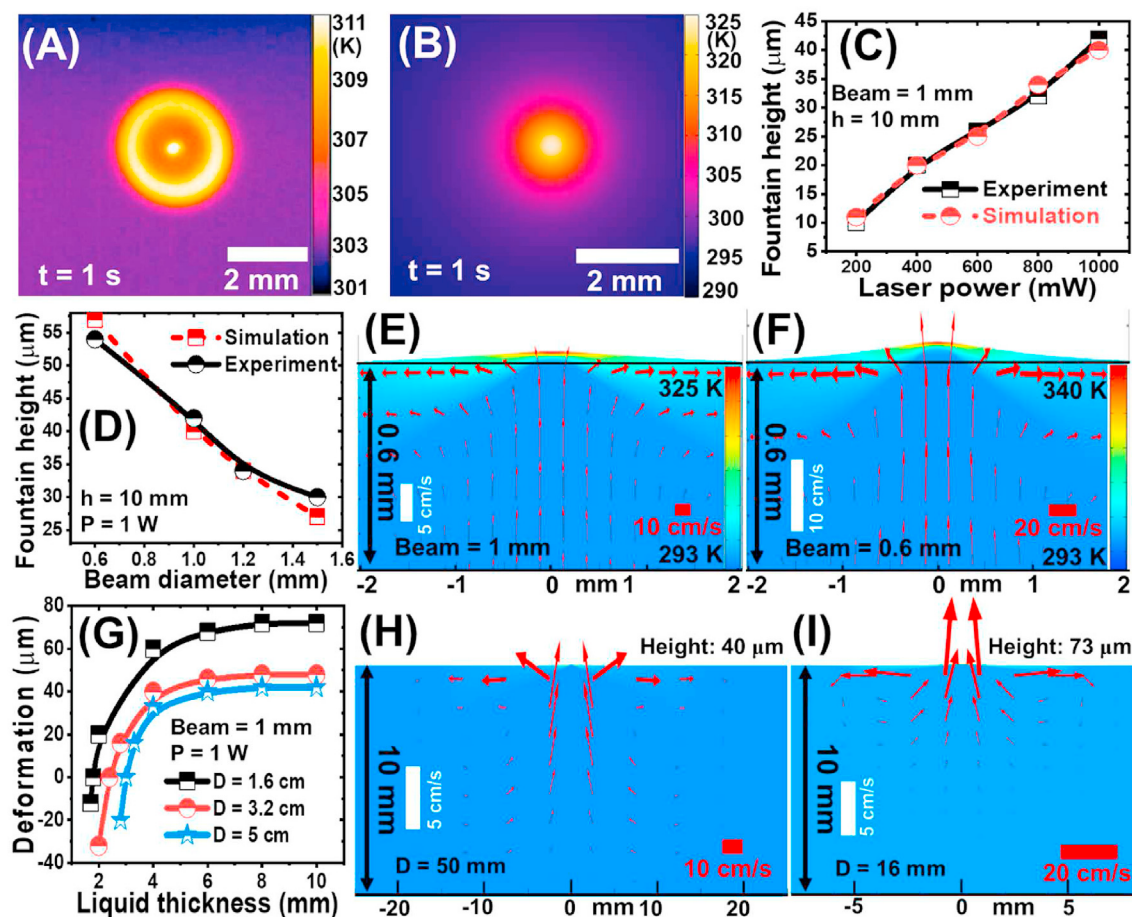


Fig. 5. Surface deformation with different incident laser power, spot size, liquid depth and container size. (A) Measured surface temperature profile by a thermal camera. (B) Simulated temperature profile. (C–D) Effects of (C) laser power and (D) beam diameter on the fountain height. (E–F) Simulated surface deformation, temperature and flow patterns with (E) 1 mm and (F) 0.6 mm laser beam diameters. The thickness of ferrofluid is 10 mm. (G) Effect of surface area and liquid thickness on the surface deformation of ferrofluid. (H–I) Simulated flow patterns with petri dish diameter of (H) 50 mm and (I) 16 mm. The laser beam diameter is 1 mm. Laser power is fixed at 1 W for all except (C).

artifact can be clearly seen by a ripple in Supplementary video S4. Because infrared camera probes average temperature below surface within an infrared penetration depth, such measured temperature is a few degrees lower than the surface temperature shown in Fig. 5B. Such a mild temperature as well as an active visible laser beam certainly will not induce any obvious damage to the ferrofluid. On the other hand, ferrofluid is not uniquely able to form laser fountain. By changing the color of inherently transparent kerosene with candle dye or rhodamine B, similar fountains can be observed in Figs. S8 and S9. In particular, Supplementary video S5 and S6 show instantaneous effect of the thickness on the surface deformation for both ferrofluid and dyed kerosene. This effect alone can immediately eliminate other competing theories for laser fountain such as radiation pressure [35], laser induced chemical change [36], inhomogeneity or imperfectness of the ferrofluid, and certainly cannot be understood from static interface force balance which has been used to explain liquid-solid contact angle or meniscus.

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Having established Marangoni convection as the driving force

for the laser fountain, we can systematically explore the effect of laser power, the size of laser beam and liquid area on laser fountain. As expected, Fig. 5C shows that the fountain height is nearly proportional to the laser power. This is understandable since a higher laser power will create a larger temperature gradient and thus a stronger Marangoni convection. Due to the same reason, Fig. 5D–F and Fig. S10 show that a focused laser beam will generate a stronger fountain, resulting in a lower critical thickness at which the net elevation is zero with a cone-in-depression as shown in Fig. 2. Fig. 5E also shows that the simulated surface flow speed is very close to the measured value from Supplementary video S1 in our typical experiment with a laser power of 1 W and diameter of 1 mm. Surprisingly, the surface area of the liquid also has a significant effect. Fig. 5G reveals that fountain becomes much taller and the critical thickness become smaller when the diameter of surface is reduced from 5 cm to 1.6 cm. To understand this effect, we again performed the simulation. The results in Fig. 5H and I have not only correctly predicted the fountain height, but also provide a physical explanation: with a large surface area, Marangoni flow spreads out freely on the surface with very little coupling to the upward recirculation flow; while in a smaller petri dish, lateral surface flow is forced to turn downward by the side wall and becomes a part of the recirculation flow. In other word, the momentum of outward surface flow is recycled to strengthen the recirculation flow, leading to a much stronger laser fountain. This boundary effect further confirms that the laser-induced fountain is

a result of dynamic fluidic circulation driven by Marangoni flow.

3. Conclusion

In conclusion, we have created laser fountains with various shapes and proved through systematic experiments and CFD simulations that laser fountains are generated by upward recirculation flow, which is driven by surface outward Marangoni convection. We have also discovered that the size of both laser beam and liquid surface can affect the fountains, and especially a smaller surface area will greatly enhance the fountain and reduce the critical thickness, however, it is still difficult to provide a simple analytic formula to predict these values. Since the Marangoni effect happens in any liquid with an arbitrary depth, our discoveries, experimental design, and simulation techniques have unraveled a previously limited understanding of Marangoni convection-induced interface phenomena and enabled us to explore new fluidic regimes from thin liquid films to any liquid with an arbitrary depth. Ferrofluid is not a unique liquid for Marangoni convection; as demonstrated above, new liquids with strong optical absorption can be achieved by varying the laser wavelength or adding light absorbing elements such as colorants or plasmonic nanoparticles [18,32]. Besides fundamental research, the Marangoni effect has already found enormous applications in nearly all branches of engineering due to the ubiquitous involvement of liquids [19,28–30]. The discovery of laser-controlled rapid and strong surface deformations through Marangoni effect is expected to open new opportunities for novel applications in surface wave generation [20], 3D printing [24], mass transport [19,36], dynamic grating and spatial light modulator [27], and adaptive optics [29].

Credit author statement

F. L., Z. M. W., J. S. and J. B. designed the experiment. F. L., A. Q., T. T., G. Y., Y. Q., T. V., J. Z., H. Zhang, H. Zhong, C. Z., P. Y., and X. T. performed the experiment. R. J., W. Z., M. M., F. L., D. L., and J. H. performed the simulation. F. L., P. Y., X. T., S. D., D. L., J. H., J. S., J. B. and Z. M. W. analyzed the results. F. L. and J.B. wrote the paper with the inputs of all authors. Z. M. W., J. S., and J. B. supervised this project.

Data and materials availability

All data is available in the main text or the supplementary materials.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.mtphys.2021.100558>.

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