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Unconventional locomotion of liquid metal droplets driven by magnetic fields

Abstract

The locomotion of liquid metal droplets enables enormous potential for realizing various applications in microelectromechanical systems (MEMSs), biomimetics, and microfluidics. However, current techniques for actuating liquid metal droplets are either associated with intense electrochemical reactions or require modification of their physical properties by coating/mixing them with other materials. These methods either generate gas bubbles or compromise the stability and liquidity of the liquid metal. Here, we introduce an innovative method for controlling the locomotion of liquid metal droplets using Lorentz force induced by magnetic fields. Remarkably, utilizing a magnetic field to induce actuation avoids the generation of gas bubbles in comparison to the method of forming a surface tension gradient on the liquid metal using electrochemistry. In addition, the use of Lorentz force avoids the need of mixing liquid metals with ferromagnetic materials, which may compromise the liquidity of liquid metals. Most importantly, we discover that the existence of a slip layer for liquid metal droplets distinguishes their actuation behaviors from solid metallic spheres. We investigate the parameters affecting the actuation behavior of liquid metal droplets and explore the science behind its operation. We further conducted a series of proof-of-concept experiments to verify the controllability of our method for actuating liquid metal droplets. As such, we believe that the presented technique represents a significant advance in comparison to reported actuation methods for liquid metals, and possesses the potential to be readily adapted by other systems to advance the fields of MEMS actuation and soft robotics.

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Unconventional Locomotion of Liquid Metal Droplets

Driven by Magnetic Fields

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Abstract

The locomotion of liquid metal droplets enables enormous potential for realizing various applications in microelectromechanical systems (MEMS), biomimetics, and microfluidics. However, current techniques for actuating liquid metal droplets either associate with intense electrochemical reactions or require the modification of physical properties by coating/mixing them with other materials. These methods either generate gas bubbles or compromise the stability and liquidity of the liquid metal. Here, we introduce an innovative method for controlling the locomotion of liquid metal droplets using Lorentz force induced by magnetic fields. Remarkably, utilizing magnetic field to induce actuation avoids the generation of gas bubbles in comparison to the method of forming surface tension gradient on liquid metal using electrochemistry. In addition, the use of Lorentz force avoids the need of mixing liquid metal with ferromagnetic materials, which may compromise the liquidity of liquid metal. Most importantly, we discover that the existence of a slip layer for liquid metal droplets distinguishes its actuation behaviors from solid metallic spheres. We investigate the parameters affecting the actuation behavior of liquid metal droplets and explore the science behind its operation. We further conducted a series of proof-of-concept experiments to verify the controllability of our method for actuating liquid metal droplets. As such, we believe that the presented technique represents a significant advance in comparison to reported actuation methods for liquid metal, and possesses the potential to be readily adapted by other systems to advance the fields of MEMS actuation and soft robotics.

1. Introduction

Gallium and several of its alloys are liquid metals at or near room temperature.¹⁻³ They possess many remarkable properties such as high electrical and thermal conductivity, large surface tension, low vapor pressure, and low toxicity compared with mercury.^{4, 5} These properties have made them very useful in various applications such as flexible electronics,^{1, 3, 6} microfluidic actuators,^{7, 8} as well as forming 3D structures.^{3, 9-11} Inspired by the flexibility and controllable surface properties of such liquid metal alloy, fracture, motion, and deformation characteristics of liquid metal droplets in electrolytes have recently become a focal point of research with several discoveries reported.^{2, 12-20} As recently discovered, a liquid metal droplet can work as a self-propulsion motor which can pave the path without human intervention;^{12, 21} a droplet immersed in electrolyte solution can be manipulated to move between a pair of electrodes by introducing asymmetrical surface tension;^{14-16, 22-26} also the induced Marangoni flow at the surface of a liquid metal droplet upon the application of a potential gradient was explored and employed to make microactuators.^{2, 27-30} Further research has shown that the liquid metal marbles, coated nanoparticles, could be propelled by generating bubbles through the photochemical reaction.³¹⁻³³ However, the abovementioned methods for controlling the liquid metal often rely on intense chemical reaction and therefore, inevitably generate gas bubbles. This could be problematic when actuating liquid metal in an enclosed system. Conventionally, magnetic fields can also be utilized to actuate liquid metal droplets in a contactless manner after coating/mixing them with ferromagnetic particles such as nickel and iron,^{34, 35} nonetheless, the liquidity and long-term stability of the liquid metal alloy can be compromised due to the formation of a new alloy between the liquid metal and the ferromagnetic particles.³⁶

As such, we have been impelled to explore new methods for actuating liquid metal droplets that are simple, chemical reaction-free, and without compromising the intrinsic properties of liquid metal. Here, we report the locomotion of EGaIn (75% gallium and 25% indium) liquid metal droplets purely driven by

magnetic fields without using ferromagnetic particles. The driving force comes from the Lorentz force derived from the eddy current, which is generated by the relative movement of the liquid metal droplets and magnetic fields. Most importantly, we discovered that the actuation behavior of liquid metal droplets is unconventional due to the presence of a slip layer, which is different from the phenomenon observed for solid metal spheres. We developed and validated the theory behind the new driving method, and conducted proof-of-concept experiments to demonstrate the viability of approach and explore the science behind its operation.

2. Experimental Section

Experimental Setup: See Supporting Information S1.

Activation of the Locomotion: The DC motor (Leadshine 57HS09) was controlled using a motor control unit (MCU, Arduino Carduino UNO R3). Permanent magnets with a magnetic flux density of ~5.5 kGs in the center were used to activate the actuation. All the experiments were conducted at room temperature (~24 °C) except for the case when investigating the effect of temperature on the locomotion performance; we varied the temperature from 15 to 35 °C in this set of experiments.

Materials: EGaIn liquid metal was purchase from Sigma Aldrich, USA. Gallium (Santech Marerials Co. Ltd) sphere was fabricated by casting liquid gallium (~50 °C) using a mold printed using a 3D printer (Formlabs Form2). Next, the mold was placed into a fridge (-10 °C), and cooled for 1 h.

EGaIn Droplet Oxidizing and Reducing Experiment: A piece of circular stainless steel plate (thickness of 0.1 mm) was attached to the bottom of a cut-through polymethyl methacrylate (PMMA) channel and sealed with hot-melt adhesive to prevent leakage. A copper pad was attached to the bottom of the stainless steel plate, and a copper electrode was inserted into the solution. A DC voltage was applied between the

electrode and the stainless steel plate using a DC power supply (RIGOL DP832).

3. Results and Discussions

The mechanism of actuation is shown in **Figure 1A**, in which we hypothesize that an EGaIn droplet (diameter < 5 mm) can be actuated after introducing eddy current by moving a permanent magnet under the droplet within an aqueous solution. The EGaIn droplet is approximated as a sphere due to its high surface tension. It is difficult to directly analyze the eddy current formed within the EGaIn droplet in the rotating magnetic field. Therefore, we simplified the model by equating the liquid metal sphere into three parallel coils (Figure 1A). When a permanent magnet approaches the EGaIn droplet, the magnetic flux density $\mathbf{B}$ experienced by the droplet increases and induces current $\mathbf{j}$ within the coils, which can be expressed as:

$$\mathbf{j} = \frac{d(\mathbf{B} \cdot \mathbf{S})}{Rdt} \quad (1)$$

where S is the area of the equivalent coil, R is the resistance of the equivalent coil. According to the Lenz's law, the current induced in the coils creates a magnetic field to oppose the change that produced it, indicating that the magnet is subjected to an obstructing force generated by the induced current when there is a velocity difference between the magnet and the droplet. According to Newton's third law, when the induced current impedes the movement of magnets, the magnets exert pulling forces $\mathbf{F}_1, \mathbf{F}_2, \mathbf{F}_3$ on the equivalent coils shown in Figure 1A. The force exerted on the droplet decreases along the Z axis as $\mathbf{B}$, and such a distribution of forces can be equivalent to a horizontal force $\mathbf{F}_e$ and a counterclockwise torque $\mathbf{M}_e$, as shown in Figure 1B. The resulting force and torque will induce the rolling of a solid metal sphere, making it travel towards the opposite direction of the moving magnet,^{37, 38} as shown in Figure 1C. Interestingly, unlike a solid metal sphere, there is a slip layer existed between the solid-liquid interface for

liquid metal placed in a solution.³⁹⁻⁴¹ Such a slip layer can act as a lubricant to prevent the rolling of the droplet and consequently, the horizontal force F_e will induce the actuation of EGaIn droplet towards the travelling direction of the magnet (Figure 1C).³⁹⁻⁴¹

To examine the hypothesis and investigating the actuating behavior, we established an actuating platform consisting of a circular PMMA channel with a semi-circular cross section groove, and permanent magnets driven by a DC motor travelling underneath the PMMA channel, as shown in Figure 1D. The detailed experimental setup is given in Supporting Information S1. Figure 1E shows the magnetic field induced actuation of an EGaIn droplet within the PMMA channel filled with 0.5 mol/L sodium hydroxide (NaOH) solution and magnetic flux density of ~ 2.3 kGs upon the activation of the DC motor at 625 RPM. The EGaIn droplet actuated along the rotating direction of the magnets (counterclockwise) at a speed of ~ 36.6 mm/s (also see Movie S1, Part 1), which aligns with our hypothesis given in Figure 1C. In order to rule out the factors contributed by the surface tension induced actuation,^{2, 24, 27, 42} we also conducted an experiment within a hydrochloric acid (HCl) solution and found that the actuating behaviors are similar to the case observed in NaOH solution as shown in Supporting Information S2 and Movie S1, Part2. On the contrary, we observed that a solid gallium and copper metal sphere travelled in the opposite direction (clockwise) of the rotating magnet, as shown in Figure 1F (also see Movie S2 and Supporting Information S3). This experiment further proved our theory on the different actuating behaviors for solid and liquid metal spheres.

Figure 1. Locomotion of liquid metal droplets and solid metal spheres. (A) Schematic of the forces exerted on the equivalent coils in a metallic sphere, the blue arrows indicate the direction of the magnetic field; the green arrow indicates the travelling direction of a magnet; and the black arrows indicate the forces experienced on the equivalent coils. (B) Schematic of the equivalent force and torque exerted on

the sphere. (C) Schematics showing the actuation of a solid ball and a liquid metal droplet placed on a substrate. (D) Schematic of the experimental setup. Sequential snapshots for the locomotion of (E) an EGaIn droplet (0.2 mL) and (F) a solid gallium ball (radius of 4 mm). (G) Image showing the movement of an EGaIn droplet and sequential snapshots for the self-rotation of an EGaIn droplet. (H) Cross-sectional view for an EGaIn droplet experiencing uneven distribution of magnetic field.

Interestingly, we also observed the self-rotation of the EGaIn droplet while it is travelling along the PMMA channel, as shown in Figure 1G, in which we used red particles to coat the surface of EGaIn droplet to clearly show the rotation (also see Movie S3). We believe that the self-rotation can be attributed to the fact that the magnetic flux density experienced by the two hemispheres of EGaIn droplet is uneven due to the imperfect alignment when a magnet passed under the EGaIn droplet, as shown in the cross-sectional schematic given in Figure 1H. Such a misalignment will induce a larger Lorentz force on one side of the hemisphere and generate a torque in the horizontal plane to rotate the droplet. The mechanism for explaining the self-rotation phenomenon was further validated by conducting experiments using channels with different configurations to induce different asymmetry scenarios of magnetic fields on the EGaIn droplet, as discussed in Supporting Information S4.

Upon the successful demonstration of the magnetic field induced actuation of liquid metal droplet, we conducted a series of experiments to investigate the performance of locomotion as a function of motor speed, the sizes of the EGaIn droplets, the concentration of NaOH solution, and the magnetic flux density. **Figure 2A** shows that a faster actuation speed can be achieved by increasing the rotating speed of the DC motor. This is due to the fact that a higher motor speed can induce larger rate of magnetic flux change and therefore, induce a larger current which eventually translates into a bigger driving force $\mathbf{F}_e$ and torque $\mathbf{M}_e$ (see Equation 1). Interestingly, we found that by increasing the size of EGaIn droplet a higher actuation

speed can be achieved, and a lower rotating speed of DC motor is required to actuate the droplet (Figure 2A). This is probably due to the enlarged driving force caused by the increase of S and the decrease of R in a larger droplet, and the presence the slip layer minimizes the effect of friction between EGaIn and the substrate.

Next, we investigated the effect of NaOH concentration on the locomotion speed of the droplets, as shown in Figure 2B. We found that for smaller EGaIn droplets (0.1 and 0.2 mL) the actuating speed increased when using NaOH solution with a higher concentration. However, no further increase of speed was observed for the results obtained using NaOH solution with a concentration higher than 0.3 mol/L. We found that the presence of NaOH or HCl is necessary for removing the oxide layer to minimize the friction, as no locomotion was observed when using deionized (DI) water. For the case of 0.3 mL droplet, locomotion speed is independent of the NaOH concentration. We believe that increasing the concentration of NaOH prevents the oxidation of the liquid metal, which is beneficial for driving the liquid metal droplet.

We further studied the effect of the magnet field on the locomotion performance by varying the magnetic flux density $\mathbf{B}$ and the number of magnets used, as shown in Figures 2C and D. It is evident that a higher $\mathbf{B}$ can lead to a larger induced force and results in a larger locomotion speed. In addition, increasing the number of magnets from 2 to 4 can elongate the time of the driving force exerted on EGaIn droplet when using the same rotating speed of the DC motor, therefore, leading to a higher locomotion speed for EGaIn droplets with the same size.

We also ruled out the effect of temperature on the performance of locomotion by conducting experiments at different temperatures ranging from 15 to 35 °C, as detailed in Supporting Information S5, in which we did not observe obvious change of actuating speed at different temperatures. Moreover, we further investigated the long-term performance of locomotion for the EGaIn droplet, as shown in Figure S6. We discovered that the actuating speed is relatively stable in the first 10 min; however, the speed

gradually decreased to only ~50% of the original speed after 150 min. This is probably due to the fact that the EGaln droplet is not fully submerged in the NaOH solution in our experiment, and the part exposed to air can be oxidized. Consequently, the chemical reaction between the oxide layer and the NaOH solution gradually consumes NaOH and reduces its concentration. The actuating speed decreases as the NaOH solution is not able to efficiently remove the oxide layer. We found that the actuating speed can be restored after replacing the NaOH solution.

Figure 2. Characterization of the locomotion performance with respect to different parameters. (A) Plots of locomotion speed vs. speed of motor. (B) Plots of locomotion speed vs. concentration of NaOH. (C) Plots of locomotion speed vs. magnetic flux density. (D) Plots of locomotion speed vs. sizes of droplets.

After examining the parameters affecting the locomotion performance, we further demonstrated the

controllability of our method for manipulating EGaIn droplets. Based on the actuation performance characterized in Figures 2A and D for EGaIn droplets with different sizes, we firstly conducted an experiment to show the chasing and merging of EGaIn droplets with different sizes, as shown in **Figure 3A**. Three droplets with the volumes of 0.1, 0.1, and 0.18 mL were placed into the PMMA channel filled with 0.5 mol/L NaOH solution at different locations, and actuation of the droplets occurred upon the activation of the DC motor at 625 RPM. Interestingly, the 0.18 mL droplet moved faster than the other two droplets, and as a result, the 0.18 mL droplet caught up with the 0.1 mL droplet in the front after ~ 8 s, enabling merging to form a bigger droplet. The merged 0.28 mL droplet actuated at an even faster speed (~ 55 mm/s) that caught up and eventually merged with the 0.1 mL droplet only after ~ 4.0 s. This chasing and merging process is also clearly shown in Movie S4.

Figure 3. Controlling the motion of EGaIn droplets. (A) Sequential snapshots of the chasing of three liquid metal. (B) Schematic of the experimental setup for the electrochemical control of EGaIn droplets. (C) Snapshots of the locomotion after oxidizing and reducing the surface of the liquid metal.

From our experiments we learnt that the presence of a solid oxide layer on the surface of EGaIn is able

to significantly compromise or even prevent the locomotion by thinning the slip layer and increasing the friction. Therefore, we hypothesize that the magnetic field induced locomotion of an EGaIn droplet can be controlled by oxidizing or reducing its surface using electrochemistry. In doing so, we placed a cut-through PMMA channel on top of a stainless steel sheet, and submerged a copper electrode into the 1 mol/L NaOH solution without touching the EGaIn droplet. The schematic of the experimental setup is given in Figure 3B, in which we can see that the EGaIn droplet can be electrochemically oxidized or reduced by applying a potential between the stainless steel sheet and the copper electrode. We observed the locomotion of the EGaIn droplet in the beginning after activating the DC motor at 625 RPM; interestingly, the actuation of the EGaIn droplet was immediately ceased and we observed the elongation of the droplet as soon as applying a 7.5 V oxidizing potential to the stainless steel sheet, as shown in Figure 3C. The elongation of the droplet indicates the oxidation of the liquid metal as the oxide layer significantly lowered its surface tension.^{15, 43-46} Locomotion of the droplet was restored after we change the polarity of the potential to electrochemically reduce the liquid metal (Figure 3C). This experiment clearly demonstrates that the motion of EGaIn droplets can be flexibly controlled by altering the property of their surface (also see Movie S5).

4. Conclusions

In summary, we demonstrated a novel method for controlling the locomotion of liquid metal droplets by purely using magnetic fields without introducing ferromagnetic particles. The driving force comes from the Lorentz force derived from the eddy current, which is generated by the relative movement of the liquid metal droplets and magnetic fields. We discovered that the actuation behaviors of liquid metal droplets are unconventional and distinct from the case observed for solid metal spheres due to the presence of a slip layer, where an opposite locomotion direction was induced for the droplet and solid sphere by the same magnetic field. Moreover, the locomotion speed of the droplet can be easily tuned by varying the size of the droplet, the concentration of the NaOH solution, and the magnetic flux density. Consequently, the liquid metal droplets with different sizes can exhibit chasing and merging behavior in one channel actuated by magnetic fields. Most importantly, the actuation of the droplets can be controlled by forming/removing the solid oxide layer electrochemically on the surface of EGaIn to thin/thicken the slip layer. The developed method for actuating liquid metal droplets is simple, chemical reaction-free, and without compromising the intrinsic properties of liquid metal. Nonetheless, it is worth noting that current platform still requires relatively bulky rotating magnets and motor; however, we believe this problem can be resolved by introducing programmed electromagnetic fields in our future works. As such, the novel phenomenon, combined with unique properties of liquid metal droplets, possess the potential to enable an exciting platform for achieving applications that cannot be realized using solid metal spheres for soft robotics, as well as the assembly of multi-functional MEMS and actuators.

Conflicts of interest

There are no conflicts of interest to declare.

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