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Tunable Superparamagnetic Ring (tSPRing) for Droplet Manipulation

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The manipulation of droplets via a magnetic field forms the basis of a fascinating technology that is currently in development. Often, the movement of droplets with magnets involves adding magnetic particles in or around the droplet; alternatively, magneto responsive surfaces may also be used. This work, presents and characterizes experimentally the formation and properties of a tunable superparamagnetic ring (tSPRing), which precisely adjusts itself around a water droplet, due to liquid–liquid interaction, and enables the physical manipulation of droplets. The ring is made of an oil-based ferrofluid, a stable suspension of ferromagnetic particles in an oily phase. It appears spontaneously due to the oil–water interfacial interaction under the influence of a magnetic field. The ferrofluid–water interaction resembles a cupcake assembly, with the surrounding ring only at the base of the droplet. The ring is analogous to a soft matter ring magnet, showing dipole repulsive forces, which stabilizes the droplets on a surface. It enables robust, controllable, and programmable manipulation of enclosed water droplets. This work opens the door to new applications in open surface upside or upside-down microfluidics and lays the groundwork for new studies on tunable interfaces between two immiscible liquids.

1. Introduction

Droplet manipulation is gaining great interest in various fields, including technological applications and fundamental studies in dynamic systems.[1,2] Special interest is adopted by the Lab-on-a-chip and microfluidics community, interested in the precise manipulation of small volumes of fluids, droplet microfluidics.[3–5] Non-invasive displacement of droplets on surfaces has been already achieved by different actuation mechanisms triggered by magnetic fields, (magneto manipulation),[6–9] electrical signals (electrowetting),[10–13] light,[14,15] chemical reactions,[16,17] sound,[18,19] and vibration.[20] The most successful method to control the movement of several droplets on a dielectric surface for microfluidics applications is electro-wetting. Some disadvantages of this approach are the need of specific type of substrate, complex fabrication process, and limited compatibility with biological samples since it applies an electrical field into the droplet. Instead, the magnetic manipulation of droplets, driven by an invisible and remote magnetic field, is harmless in a big extend for biological samples.[21] Up to date, magnetic manipulation commonly relies on loading magnetic particles inside[22,23] or outside of the droplet to move (magnetic liquid marbles and cloaked droplet),[24–27]) or on magneto responsive surfaces.[28] Ferrofluids have been already used as magnetic soft substrates[29–33] to induce a change in the interaction between droplets and substrates enabling droplet manipulation. Despite all the progress in droplet microfluidics, a number of challenges remain in the field such as its applicability for wide range of droplet volumens and the possibility to avoid fabrication of complex substrates. Current systems can mainly handle large droplets and manipulation is highly dependent on the interaction between the substrate and the droplet.

Ferrofluid is a magnetic liquid that is formed by stable colloidal suspensions of iron nano particles in a carrier medium such as oil or water. Under the influence of a magnetic field, it experiences Rosensweig instability and creates a regular self-organized pattern seen macroscopically as peaks and valleys.[34,35]

We have observed that the introduction of a water droplet in a Rosensweig instability pattern spontaneously forms a ferrofluid ring around the water droplet that remains integrated

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within the regular pattern of instabilities. This ring surrounds the base of the water droplet all along its perimeter and behaves like a magnetic instability, allowing the droplet to stabilize and separate from other droplets.

In this manuscript, we report the formation and characterization of a new type of instability, the tunable superparamagnetic ring (tSPRing), as well as whether this phenomenon could be exploited for the programmable magnetic manipulation of non-magnetic optically addressable droplets. Our experimental observations indicated that the interaction of ferrofluid instabilities with other liquids can form the basis of a very versatile and robust methodology for handling droplets on open surfaces using magnetic fields. Likewise, the manipulation of droplets through soft materials such as ferrofluids gives insights for new fundamental studies on the interaction between immiscible liquids.

2. Results and Discussion

2.1. Formation of Ferrofluid Ring

Suspension of nano-sized iron particles in ferrofluid response to a magnetic field (alignment of magnetic moments) disappears once the field is removed.[36] Up to a certain magnetic field strength (saturation magnetization, M_s), a ferrofluid stays as a continuous liquid phase. However, when the field strength overpass the saturation level, it undergoes Rosensweig instabilities[37] and self organizes as an ordered spike pattern (**Figure 1**A). The size and interspace distance of the spikes in that pattern is related to the viscosity of the ferrofluid, field strength (H) and field curvature of the magnetic field.[38] By turning on and off the magnetic field it is possible to switch between the continuous liquid phase and the discontinuous spike pattern phase.

The interaction of oil-based ferrofluid with a polar liquid such as water, is like a conventional oil–water interface which is widely studied and described in literature.[39,40] In the absence of magnetic field, when a water droplet comes in contact with a layer of ferrofluid (deposited on a surface), the ferrofluid tends to envelop the droplet due to the capillary phenomena to minimize the interfacial tension. But, would this effect be valid for a water droplet in contact with ferrofluid under a magnetic field, when it is already forming a spike pattern? In order to study that, we performed several experiments, first we placed a water droplet inside a ferrofluid static spike pattern. On a different experiment, the water droplet was placed on top of a continuous layer of ferrofluid, and after the magnetic field was applied.

Finally, ferrofluid was slowly added dropwise between two droplets of water placed on a surface on top of a permanent magnet. Figure 1A shows the result of introducing a water droplet in between ferrofluid spikes. The spikes touching the water droplet turned into a continuous liquid medium around the droplet, forming a ring that could be observed macroscopically. In the second experiment, the ferrofluid covered the whole water droplet in the absence of magnetic field, once the field was applied, the ferrofluid shaped to form instabilities, including the ring around the water droplet (Figure 1B).

In the third experiment, the spike pattern dynamically formed and broadens by increasing the volume of ferrofluid that touched the surface. As soon as the spikes reached one of the water droplets, the ferrofluid surrounded the perimeter of the water droplet in less than a millisecond acquiring the form of a ring (Video S1, Supporting Information). The ferrofluid–water interaction was shaped like a cupcake, with the surrounded ring only at the base of the droplet, in contrast to the ferrofluid cloaking method in which the ferrofluid covers the whole of the droplet.[41] However, although the droplet remained translucent (it was possible to see inside), it was covered with a thin cloaking layer of oil (oil phase of the ferrofluid) (Video S2, Supporting Information).

We examined the geometry of the ring, which presented an asymmetric cross section. Its longitudinal profile was oblique to the surface at the ferrofluid–air interface but concave at the

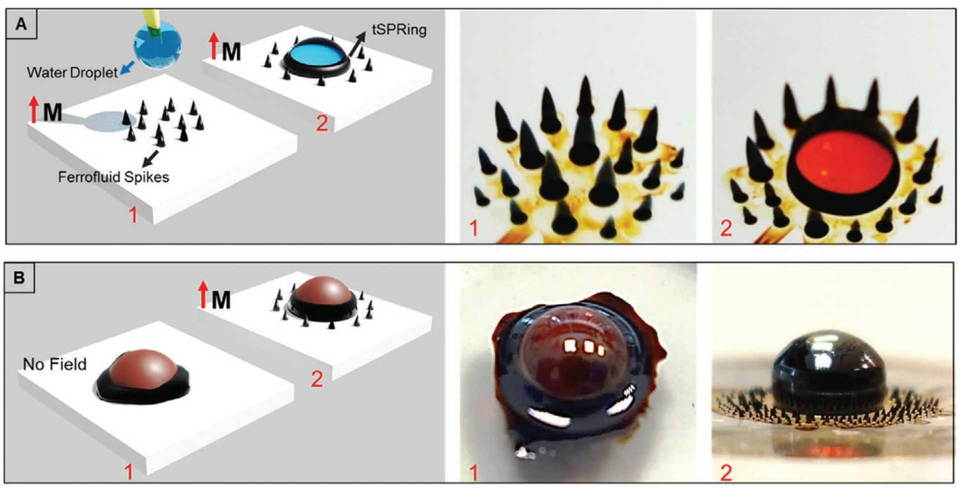

Figure 1. Formation of tSPRing by two different methods. A) A magnetic field over the M_s of the ferrofluid is applied to a layer of ferrofluid deposited over a surface, the instabilities form and the droplet of water is loaded between the instabilities. Photograph of spike pattern formed by out-of-plane magnetic field induced by a magnet positioned underneath (middle), and photograph of the tSPRing formed around a water droplet, which was pipetted in between the spikes (right). B) A water droplet loaded on continuous ferrofluid phase (deposited on top of a surface), and subsequently the magnetic field over M_s of the ferrofluid is applied. Photograph of the water droplet covered by a continuous layer of ferrofluid (middle), and photograph of the droplet after applying the magnetic field to form the instabilities (right). The tSPring forms in both cases around the water droplet as one more instability.

ferrofluid–water interface (Figure S1, Supporting Information), following the lines of the magnetic field as a magnetic instability.

The formation of the ring happened due to the introduction of an immiscible liquid in contact with the ferrofluid spike pattern which disturbed the thermodynamic equilibrium of the spikes and the system was then forced to reshape itself to find a new state of minimum energy. Since the surface tension of ferrofluid is lower than the surface tension of water, the ferrofluid changed its shape to tackle the higher tension dictated by water droplet. After the formation of the ring, the system reaches a new thermodynamic equilibrium. Regarding the asymmetric cross section, normally the spikes are in contact with two phases, the substrate and the air. When the water droplet was introduced in the pattern and touched some ferrofluid spikes, three phases were in contact with the ferrofluid, solid, liquid, and air phase, producing three different interfaces. For this reason, the ring presents an asymmetric cross-section, which is expected considering that the surface tension of the water–ferrofluid interface is lower than the surface tension of the ferrofluid–air interface.

In view to its morphology and its superparamagnetic properties, we named the enclosing ferrofluid ring as tunable superparamagnetic ring, shortly tSPRing. Below we describe a set of experiments to understand the characteristics and properties of tSPRings.

2.2. Magnetization of the tSPRing and Stability of Water Droplet

Each spike in the classical Rosensweig pattern behaves as an independent dipole, which is attracted to the center of the magnet (higher field gradient) and is repelled by the neighboring spikes. A similar behavior was expected from the tSPring. To confirm this hypothesis we pipetted several droplets of water on a PMMA surface containing ferrofluid spikes and observed the dynamic behavior of the system. A tSPRing formed immediately around each droplet, each tSPRing repelled nearby tSPRings and spikes, inducing the movement of droplets and spikes through the surface until the whole system reached an equilibrium (**Figure 2**). The obtained pattern comprised of spikes and ringed droplets followed the critical periodicity described by Rosensweig instability periodicity.[38]

The droplets that were stabilized by the rings did not spread or mix with neighboring droplets (Figure 2B,C, Video S2, Supporting Information) which confirmed that the tSPRings repel each other like the ferrofluid spikes, maintaining a fixed distance between them. Likewise, it was observed that the contact angle of the tSPRinged droplet was greater than that of the droplet without tSPRing (Figure 2D,E). This implies a smaller droplet radius, suggesting that tSPRinged droplets can support larger volumes while occupying a smaller substrate surface area. In a subsequent experiment, the distance between droplets was minimized to few microns by reducing the volume of ferrofluid (Figure S2, Supporting Information).

In view of these results we postulate that the repulsion between tSPRings and spikes was due to the magnetization of the ferrofluid in the direction of the applied magnetic field (out-of-plane), confirming that tSPRing and spikes are small soft magnets (superparamagnets with aligned magnetic domains of internally self-assembled magnetic particles in the ferrofluid) following the same magnetization axis as the external magnet underneath (Figure 2A). In other words, the ring is not only oil surrounding a droplet; it is a soft matter ring magnet that provides stability to the water droplet as long as the external field is applied.

2.3. Tunability of tSPRing

Ferrofluid is a soft material, it undergoes physical changes in the presence of a magnetic field. The response of tSPRing to fluctuations of an applied magnetic field was investigated. A 10 µL water droplet was placed on a ferrofluid layer, of 3 µL, deposited on top of a PMMA substrate. The magnetic field strength (*H*) was gradually increased by approaching a magnet, located underneath the substrate. The formation and the changes in the aspect ratio of the ring and the enclosed droplet were monitored using a video camera. In the absence of magnetic field, the droplet and the ferrofluid were relaxed over the surface with a certain degree of spreading due to the influence of interfacial tension and gravity. By introducing a 39 mT

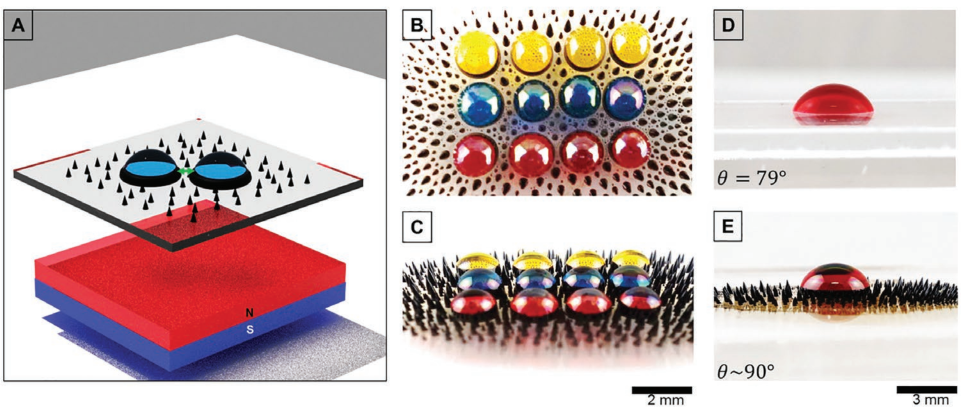

Figure 2. tSPRinged water droplets array under Rosensweig instability. A) Schematic drawing of two drops enclosed in tSPRingss that are part of the instability pattern of a ferrofluid created by the influence of a magnetic field on a PMMA surface. B,C) Photographs of a tSPring enclosed water droplets array on top of a PMMA substrate, tSPRing, and spikes showed dipole magnet behavior keeping a characteristic distance among themselves. D,E) Compare the shape and contact angle of a droplet being pipetted on clean PMMA surface and a droplet being pipetted in between ferrofluid instabilities.

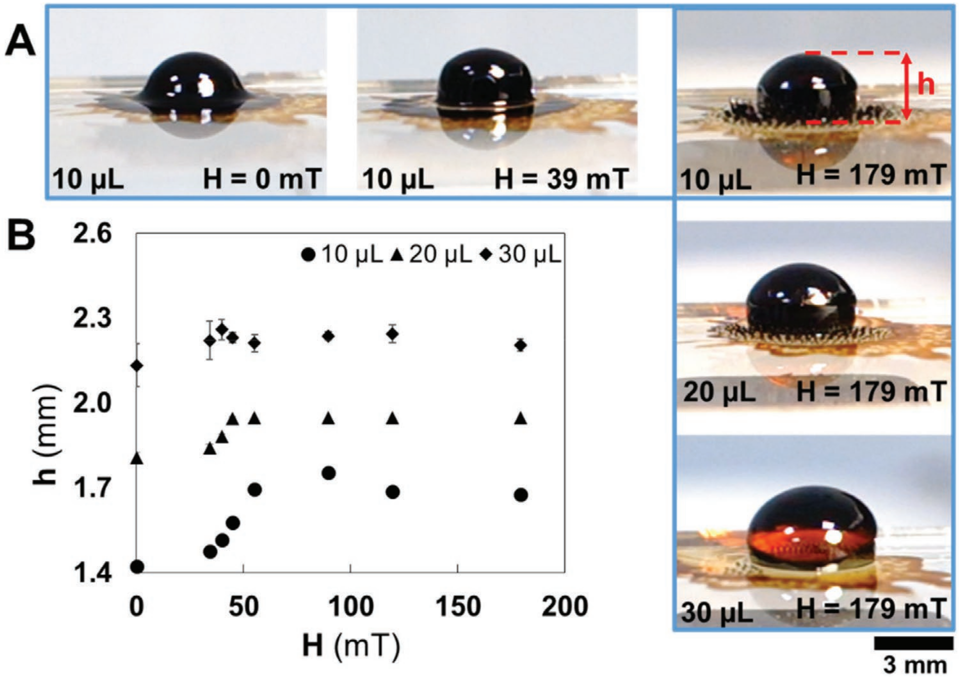

Figure 3. Aspect ratio tunability of tSPRing on up-side substrate. A) Photographs of a water droplet deposited on top of a ferrofluid film on a PMMA substrate, taken under different magnetic field strengths (horizontal series), or at constant field strength with different water-ferrofluid volume ratio (vertical series). B) Data plot of the water droplet height (h) as a function of the magnetic field strength (H) for different water-ferrofluid volume ratio (ferrofluid volume of 3 µL, droplet volumes as 10, 20, and 30 µL).

magnetic field, the spreading of the ferrofluid on the surface was reduced as it accumulated at the interface with the water droplet. The combination of droplet and ferrofluid ring started resembling a "cupcake" configuration. Further increasing of H above the saturation magnetization produced the formation of the ring with a sharp end all around the water droplet perimeter and the expected pattern of spikes around the ring. The spikes were separated from the ring keeping a characteristic distance. **Figure 3**A shows three snapshots at different H values as 0, 39, and 179 mT. The aspect ratio of the ring around the droplet changed with the magnetic field strength (H), reducing its diameter and increasing its height that forced a similar change in the morphology of the water droplet. We measured the height of the water droplet (h) versus the field magnetic strength (H), the data are plotted in Figure 3B and showed that up to the saturation of the magnetization (around 90 mT) the height of the water droplet (h) increased notably, but remained unchanged once the instabilities were formed. The increase in the height of the water droplet is related to a narrowing of the diameter of the ring (Figure S3 and Video S3, Supporting Information). The cupcake configuration was maintained the entire time while the magnetic field was on. In a subsequent experiment, we increased volume ratio between ferrofluid and water; water droplets of 20 and 30 µL were pipetted over 3 µL of ferrofluid. The ferrofluid ring around the water droplets was formed in all cases. The height of the ferrofluid ring and the slope h/H was lower for a larger water–ferrofluid ratio. While the perimeter of the water droplet was larger for larger droplet volumes, the ferrofluid volume was constant and therefore it was a limiting factor to obtain a higher ring. If more ferrofluid would be added, the ferrofluid first would be consumed to increase the height of the ring and subsequently a spike pattern would be formed.

Similar experiments were carried out on the bottom face of a PPMA substrate in order to understand if the ferrofluid ring could also effectively enclose hanging droplets. In order to obtain a hanging water droplet on the ferrofluid layer, we increased the volume of ferrofluid up to 3 µL to hold a 30 µL water droplet (**Figure** 4A). At that point, the system reached a balance between the two competing forces: the capillary force at the interface of water-ferrofluid and the gravity.

The ferrofluid and water droplet interaction in absence and presence of applied external field was similar to prior experiments in Figure 3. When the magnetic field was applied we observed again the formation of the ring around the water droplet, which underwent an increase of its aspect ratio with the strength of the magnetic field (Figure 4B), after the saturation of the magnetization we observed the formation of the spikes as in the upside surface experiment. The elongation of the droplet caused by applied external magnetic field was measured and plotted showing a similar behavior (Figure 4C).

These observations demonstrated the tunability of tSPRing as a dynamic adaptation of its aspect ratio in response to the variation of an external magnetic field. tSPRing acted as a dynamic pinning ridge for water droplets enabling the tuning of their morphology and contact angle with a solid substrate (droplet-solid substrate point).

2.4. Controllable and Programmable Manipulation of Droplet by tSPRing

The results in Section 2.2 showed that tSPRing behaves like a soft magnet, analogous to a solid dipole ring magnet. Once it has been formed, it should be possible to translocate it in a

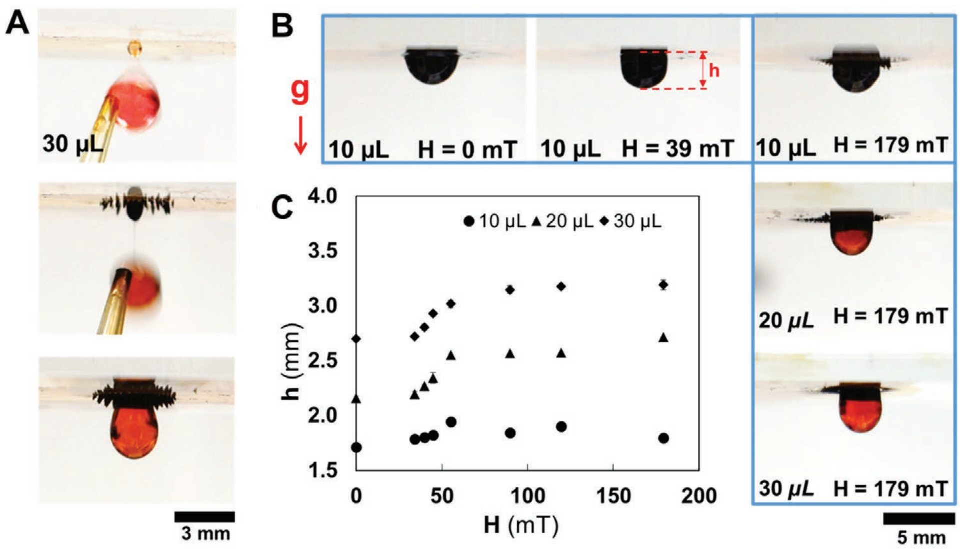

Figure 4. Aspect ratio tunabilityof tSPRing on up-side down substrate. A) Photographs of 30 µL water droplet being pipetted on a PMMA up-side down surface, wetted with ferrofluid (top), with a 1.5 µL of ferrofluid (middle), and with 3 µL of ferrofluid (bottom) under the influence of a 90 mT magnetic field. Increasing amount of ferrofluid at the upside-down surface resulted in holding the droplet against the gravity by ferrofluid-water interactions. B) Photographs of water droplets of different volumes deposited on top of a ferrofluid film on an up-side down PMMA substrate, under different magnetic field strengths (horizontal series), or at constant field strength with different water-ferrofluid volume ratio (vertical series). C) Data plot of the height (*h*) of the water droplet as a function of the magnetic field strength (*H*) for different water-ferrofluid volume ratio (ferrofluid volume of 3 µL, droplet volumes as 10, 20, and 30 µL).

controlled manner by moving the magnet on the other side of the substrate, as indicated in **Figure 5**. Our observations confirmed that the "water-ferrofluid cupcake" could be handled in a controlled manner by the movement of the magnet. Both cases, sessile and pendant droplets could be moved along the substrate through simple translocation of the ring induced by the

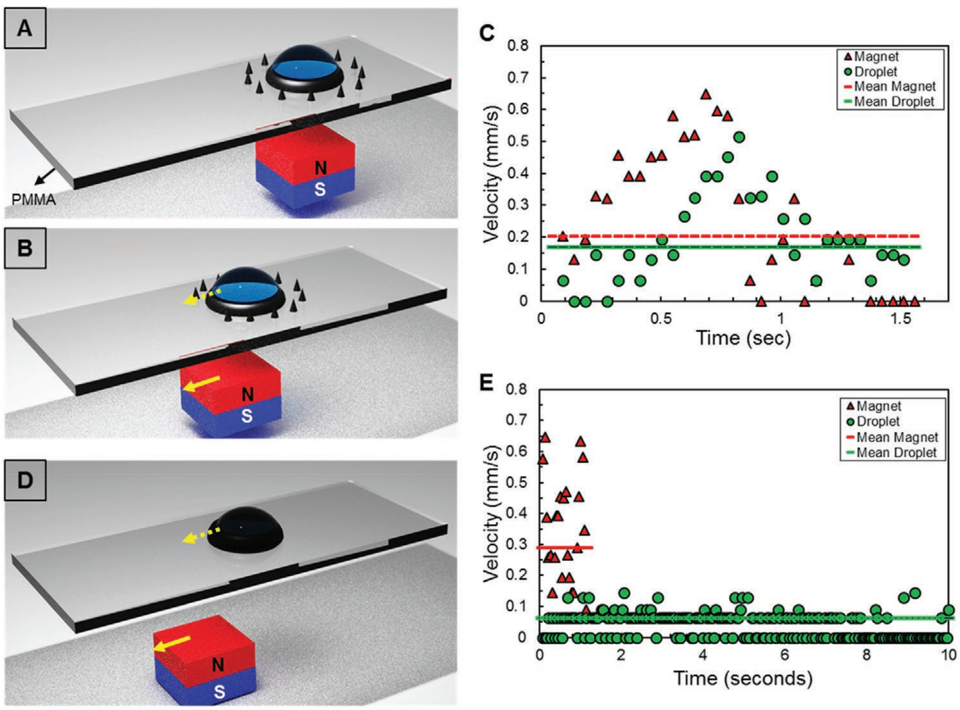

Figure 5. Translocation of water droplet by ferrofluid tSPRing. A,B) Schematic representation for translocation of tSPRinged droplet at *H* values over M_s. The movement of the magnet under the substrate forced the ferrofluid to move in the same direction. The magnetic instabilities, both the spike pattern and the ring that wrapped the water droplet moved according to the movement of the magnet. C) Plot showing the velocity of the magnet and the water droplet versus time for case (B). D) Schematic representation of the translocation of droplets wrapped in cloaking manner at *H* values below M_s. E) Plot showing the velocity of the magnet and the water droplet versus time for case (D).

movement of the permanent magnet located at the other side of the substrate (Videos S4 and S5, Supporting Information)

To compare the movement of the magnet with the movement of the water-ferrofluid assembly, the magnet underneath the substrate was moved manually and the movements of the magnet and the droplets were optically tracked. Figure 5C–E shows acceleration values of the droplet plotted against the acceleration of the magnet underneath. The graph D shows that the acceleration of the cupcake was very similar to the acceleration of the magnet in H values over M_s, which resulted in similar mean velocities. But, in the case of H values below M_s there was a considerable delay between the acceleration of the magnet and the cloaked droplet.

These results indicated that water-ferrofluid cupcakes can be robustly moved by moving the external magnetic field enabling a precise and programmable manipulation of the enclosed droplet. Moreover, relocation of the ringed droplet was as fast as the relocation of the permanent magnet. It is especially remarkable the robust translocation of pendant droplets, an unusual example.

2.5. On/Off Switchable tSPRing, Mixing of Droplets

In the previous sections of this manuscript, the formation of a ferrofluid ring and the ferrofluid-water cupcake has been described. It would be of interest to study the possibility of mixing the contents of two cupcakes (droplets) in a controlled way. To do this, we formed water-ferrofluid cupcakes with two different droplets of water. Using a small mechanical force, we joined the two cupcakes, which interacted through their ferrofluid rings, but did not mix because the two droplets of water were not in contact, physically separated by ferrofluid barrier that was formed (**Figure 6**A). By removing the magnetic field, the tSPRing began to behave like a liquid oil phase; gravity expanded it across the surface and finally allowed contact between the water droplets. In a few seconds the system was reconfigured to go from a figure of eight (resembles the shape of number eight) in which both droplets were independent, to a circular shape where the content of both droplets slowly mixed by diffusion (Figure 6C and Video S6, Supporting Information). By removing the external magnetic field, both Brownian motion of the iron colloids inside the ferrofluid and the gravity force helped the expansion of the ferrofluid on the substrate in any direction. Consequently, this allowed the water droplets to join and forced the reconfiguration of the system into a state of minimum internal energy, the circle. By turning on the field again, the later cupcake assembly could be mechanically approached and merged with another enclosed droplet and so on.

The paramagnetic nature of the ferrofluid enabled that the ring structure could be turned on and off accordingly by applying or removing the magnetic field. This enabled both, the precise movement of the droplets along the substrate and their merging when needed.

3. Conclusion

A tunable superparamgnetic ring (tSPRing) is spontaneously formed around a water droplet when an oil-based ferrofluid is in contact with the droplet under the influence of a magnetic field. The interfacial interaction between both liquids and the soft magnetic characteristics of the ring allows a robust, controllable, and programmable manipulation of the enclosed droplets. The water droplet can be precisely moved by moving the external magnetic field. The combination of the tSPRing and a water droplet, resembles the cupcake assembly. This assembly could be formed on top of a substrate, or as a hanging cupcake on the underside of a substrate. The dimension of the ring depends on the volume relationship between the water and the ferrofluid, while its aspect ratio (height to diameter) is related to the magnetic field strength and the field curvature. The ring is a paramagnetic magnet and behaves as an instability staying away from other instabilities at a defined distance. The ring

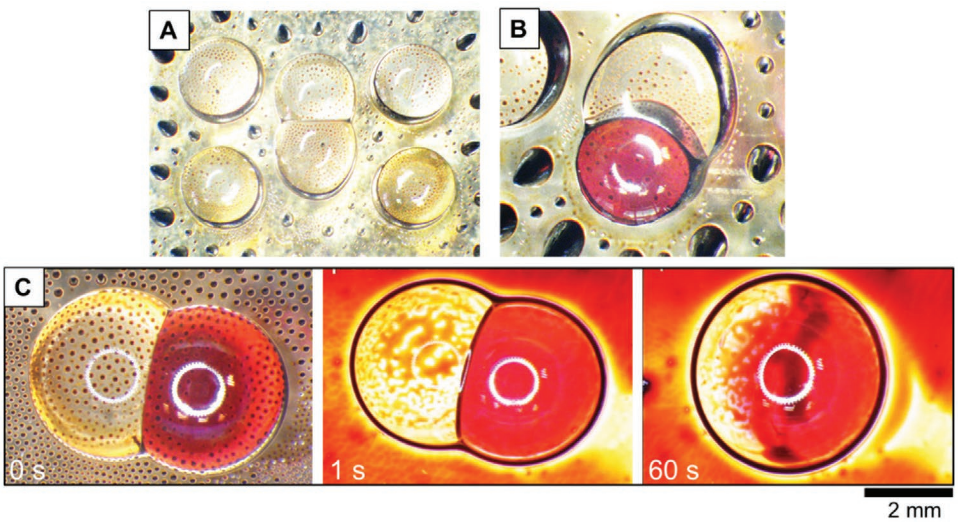

Figure 6. On/Off switchable ferrofluid ring, mixing of droplets. A,B) Photographs of two droplets merged but not mixed, under the influence of a magnetic field. C) Screenshots 1 and 60 s after removal of the magnetic field. In the absence of the magnetic field the tSPRing spreads on the surface, the physical barrier between both droplets disappear and the droplets mix spontaneously.

encloses the water droplets stabilizing them and preventing their mixing. Even when two or more cupcakes are mechanically brought together the water droplets do not mix, because their ferrofluid rings fuse to form a physical isolating barrier. However, the tSPRing is an on–off switchable structure and those droplets can be mixed by turning off the magnetic field.

In comparison with other droplet manipulation systems, tSPRing is based on liquid–liquid interfacial interaction. It allows droplet manipulation over wide range of substrates and does not require complex fabrication processes. tSPRing enables a versatile and generic fluidic control platform for droplets of any size and volume. The fact that both the liquids, the ferrofluid and the water, are immiscible prevents the contamination of the water droplets and allows the easy recovery of its contents. To the best of our knowledge, the use of tSPRing for manipulation of a hanging droplet is the first example of magnetic manipulation of droplets on an upside-down surface, what opens the door to novel applications. Likewise, it provides a new scenario for fundamental studies on oil–water interface since the external magnetic field modifies the natural capillarity wrapping of the water droplet. We conclude that tSPRing constitutes an advantageous new approach for open surface droplet microfluidics.

4. Experimental Section

Ferrofluid: The ferrofluid used for experiments was a commercial type from the oil-based series, coded EMG900 (Ferrotech USA). It had the highest saturation magnetization (99 mT) in oil-based ferrofluids with 17.7% volume of magnetic particles concentration. Among the oil-based ones, EMG900 has relatively low viscosity (60 mPa s) which allows easier formation of Rosensweig instabilities and a strong paramagnetic effect due to less viscosity which goes in favor of losing magnetic property once the field is removed by the help of Brownian motions.

Magnets and Controlling of Magnetic Field: Two permanent neodymium magnets with magnetization of 450 mT were used during the experiments (purchased from firstformagnets). The sizes were as 10 mm square cubic and 20 × 30 × 10 width × length × height rectangular cubic one in order to have bigger surface area. Both had magnetization axis on the 10 mm side. Controlling of magnetic field strength was done by reducing the distance in between the surface of magnet and the deposited ferrofluid on PMMA surface. A standard Laboratory Scissor Lift Riser was used to move the magnet upward and downward. 10 mm magnet was used for experiments of the tSPRing formation, tenability on upside and hanging droplets, as well as droplet manipulation. For the remaining experiments the rectangular cubic magnet was used. Both magnets and all experiments were done with out-of-plane magnetic field by positioning the magnet underneath of a 4 mm thick PMMA surface. For the field strength calculations a simplified formula:

$$H_x = \frac{2Ma^3}{4\pi r^3} \quad (1)$$

was used. Where, H is the field strength from a distance (r) far from the surface of magnet. M is magnetization of the magnet as 450 mT and a is the length of magnetization axis.

Image Acquiring and Analysis: Images and videos were acquired by both Nikon DI-Fi3 camera mounted on a stereomicroscope (BetaOptics-Plan1X objective) and a digital camera (Sony-Cyber-shot DSC-RX100 with max frame rate of 100 fps). Quantification of the images to measure the height and width of the droplet in Figures 3 and 4 were done by ImageJ software. The manual tracking plugin of the same software was used to track the droplet movement which gives velocity measurements in time frame.

Substrate: 4 mm of thickness PMMA (purchased from PLEXIGLAS Evonik Industries AG) was used as substrate. It was cut by CO_2 laser machine (Universal Laser Cutter System) for desired dimensions in different experiments. The surface of the PMMA was cleaned by DI water and dried prior to the experiments.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Data available on request from the authors.

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droplet manipulation, ferrofluids, magnetic particles, microfluidics

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