Electronic Supplementary Material (ESI) for Soft Matter.

Supporting Information

Shape evolution and splitting of ferrofluid droplets on a hydrophobic surface in presence of magnetic field

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S1. Variation of magnetic field

The variation of magnetic flux density (B) with gap (g) for the magnet used in our experiments is shown in Fig. S1.

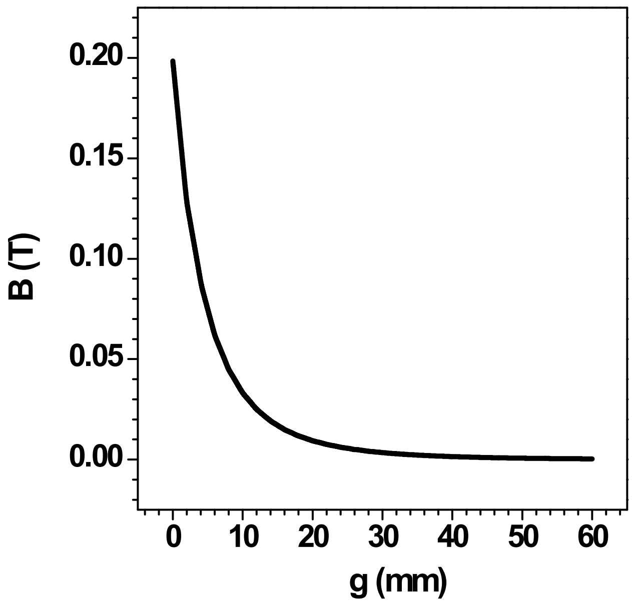
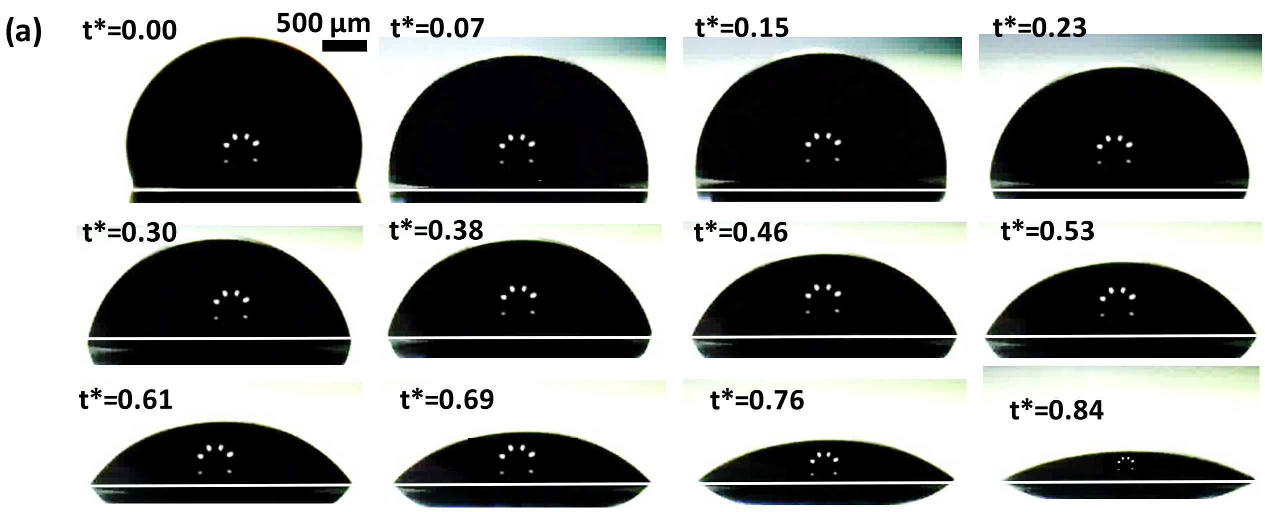


Fig. S1 Variation of magnetic field (B) with the vertical gap (g) between the magnet and the PDMS surface.

S2. Droplet evolution without magnetic field



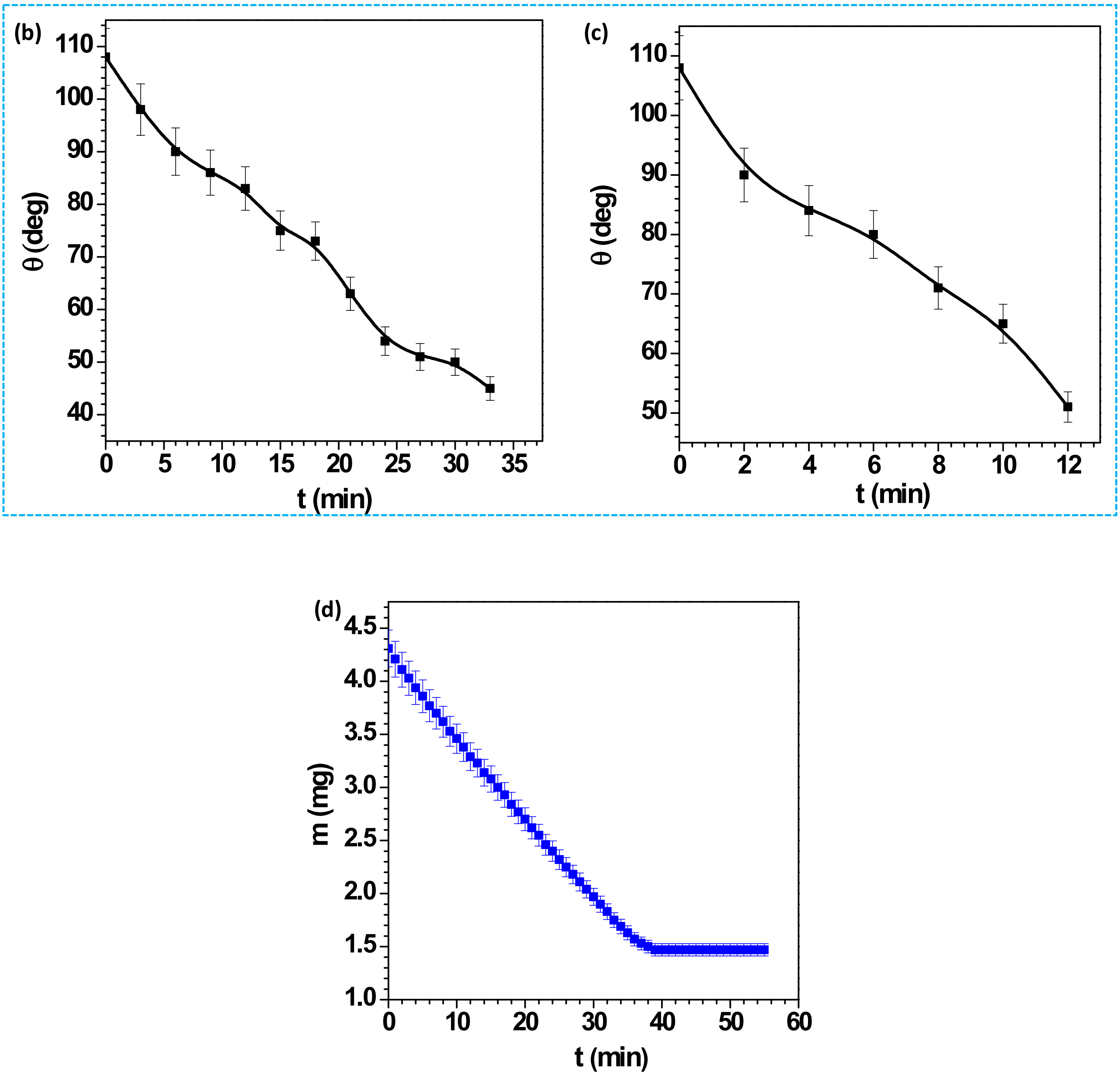


Fig. S2 (a) Image sequence of a ferrofluid droplet $(V = 4\mu L, c = 1.2\%)$ evaporating in the absence of magnetic field, (b) Variation of contact angle with time for droplet $(V = 4\mu L, c = 1.2\%)$ in the absence of magnetic field (c) Variation of contact angle with time for the same droplet $(V = 4\mu L, c = 1.2\%)$ in the presence of magnetic field $g = 1.5\ cm$ (d) Variation of mass (m) of a ferrofluid droplet $(V = 4\mu L, c = 1.2\%)$ without magnetic field with time (t).

S3. Theoretical analysis

The breaking of ferrofluid (FF) droplet in the presence of the magnet is governed by the competition between two forces - (i) Magnetic force $(F_m \propto volume)$, (ii) Surface tension force $(F_s \propto length)$. The gravitational force is not considered in this analysis since the Bond number (Bo) associated with the droplet is much less than unity. Free body diagram of the ferrofluid droplet is shown in Fig. S1. Once a droplet is placed on the PDMS coated glass slide, the magnet is brought and fixed at a particular gap on top of the droplet. The deformation of the FF droplet due to the interplay between surface tension and magnetic forces before and after interaction with the magnetic field are shown in Fig. S1(a) and S1(b) respectively. It is found that the droplet contact angle gets modified from hydrophobic to hydrophilic during the interaction. Also, the droplet height (h) and base diameter (D) follow some trend, which is discussed in the results and discussion section. We have used MATLAB 2012a to evaluate a force ratio (k) between the two major competing forces $(F_m\ and\ F_s)$ responsible for droplet breakup at a particular gap (g) between the PDMS surface and the magnet. Based on the experimental observations we have found, there exists a critical gap (g_c) at which the droplet thread breaks and sticks to the magnet, leaving a smaller droplet on the substrate.

S3.1 Magnetic force (F_m)

The magnetic force (F_m) is given by,

$$F_m = \mu_0 V \chi \nabla H^2 \tag{1}$$

where, μ_0 – vacuum magnetic permeability, χ – ferrofluid susceptibility, H – applied magnetic field due to the permanent magnet, V – ferrofluid droplet volume. Since the applied magnetic field is more than the saturation magnetization (6.6mT) of the ferrofluid, we can simplify the above equation using,

$$M = \chi H \tag{2}$$

Finally, the magnetic force is given by,

$$F_m = \mu_0 V M \nabla H \tag{3}$$

where, M – Ferrofluid magnetization. It is accounted by the well known Langevin function (L) as

$$M(H) = M_s L = M_s \left\{\coth(\gamma H) - \frac{1}{\gamma H}\right\} \tag{4}$$

where $\gamma = \frac{3\chi_0}{M_s}$, χ_0 is the initial susceptibility and M_s is the saturation magnetization of the ferrofluid. The dependence of initial susceptibility (χ_0) on several parameters is reported in literature[1], the initial susceptibility of ferrofluid varies as $\chi_o = \frac{\mu_0 n m^2}{3 k_B T}$, where, μ_0, n, m, represents the permeability of vacuum, particle number density, magnetic moment respectively and $k_B T$ denotes the thermal energy.

As found in literature[2], the variation of magnetic fields H_x, H_y for a single rectangular permanent magnet of width $2a$ and height $2b$ is found be as follows,

$$H_x(x,y) = \frac{M_s}{4\pi}\left\{\ln\left[\frac{(x+a)^2 + (y-b)^2}{(x+a)^2 + (y+b)^2}\right] - ln\left[\frac{(x-a)^2 + (y-b)^2}{(x-a)^2 + (y+b)^2}\right]\right\} \tag{5}$$

$$H_y(x,y) = \frac{M_s}{2\pi}\left\{\tan^{-1}\left[\frac{2b(x+a)}{(x+a)^2 + y^2 - b^2}\right] - \tan^{-1}\left[\frac{2b(x-a)}{(x-a)^2 + y^2 - b^2}\right]\right\} \tag{6}$$

where, M_s – residual magnetization of the permanent magnet.

S3.2 Surface tension force (F_s)

The force due to surface tension (F_s) is given by,

$$F_s = \sigma D \tag{7}$$

Where, σ – Surface tension of ferrofluid, D – Base diameter of the ferrofluid droplet.

The net force (F) on the ferrofluid droplet along Y - direction can be written as –

$$F_y = F_{m,y} + F_{s,y} \tag{8}$$

$$F_{s,y} = F_s \sin\theta \tag{9}$$

The force ratio is defined as –

$$k = \frac{F_{m,y}}{F_{s,y}} \tag{10}$$

The surface tension force tends to hold the surface in an equilibrium shape. Due to the magnetic interaction, the FF nanoparticles start to accumulate at the dropelt apex and get pulled toward the magnet due to which the droplet height (h) increases, in effect to that the effective gap (g) between the droplet and the magnet reduces. As the droplet apex approaches the magnet, the magnetic force increases, the contact angle (θ) decreases which reduces the surface tension force component $F_{s,y}$. As a result of which, the magnetic force overcomes the surface tension and splitting occurs. In the case, when the magnet is positioned at or below the critical gap (g_c) the droplet splitting is observed.

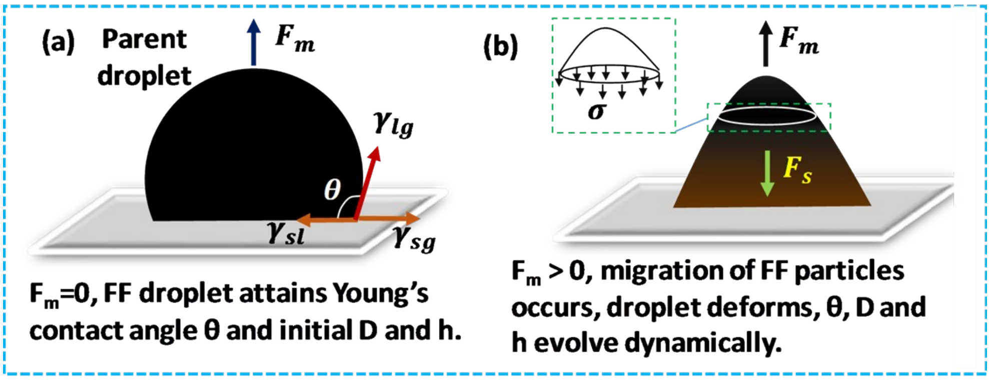


Fig. S3 (a) Schematic of the ferrofluid droplet before magnetic interaction (b) Dynamic evolution of the FF droplet when it interacts with the magnetic field before splitting.

S4. Calculation of residual droplet volume

The volume of the residual droplet is estimated using spherical cap geometry equation[3] as follows –

$$V_r = \pi D^3 \frac{(cos^3\theta - 3cos\theta + 2)}{24Sin^3\theta} \tag{11}$$

(1) Skumiel, A.; Hornowski, T. The Influence of the Concentration of Ferroparticles in a Ferrofluid on Its. **2003**, *36*, 3120–3124.

(2) Furlani, E. P. Analysis of Particle Transport in a Magnetophoretic Microsystem. *J. Appl. Phys.* **2006**, *99* (2).

(3) Nguyen, T. A. H.; Nguyen, A. V.; Hampton, M. A.; Xu, Z. P.; Huang, L.; Rudolph, V. Theoretical and Experimental Analysis of Droplet Evaporation on Solid Surfaces. *Chem. Eng. Sci.* **2012**, *69* (1), 522–529.