Breakup of a pendant magnetic drop

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We report experiments on a millimeter-sized pendant drop of ferrofluid in a horizontal magnetic field. The initial drop size is chosen just below the breakup threshold under gravity. As the magnetic field is increased, the drop tilts in order to align with the direction of the total volume force that is exerted on it: weight plus magnetic force. The breakup is controlled by a generalized Bond number based on this total force and on the radius of the neck of the drop. The evolution of drop shape turns out to be a complex process governed by many parameters such as the angle between the total force and the needle, the drop size relative to the needle radius, and the wettability of the liquid on the needle material. This suggests a certain universality, that a single value of the critical Bond number is found regardless of magnetic fluid properties and whether the force is inclined or not.

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I. INTRODUCTION

A large number of microfluidic systems are based on the generation and the displacement of droplets [1]. An external force field is generally used for the droplet manipulation. For instance, an electric or a temperature field can induce a surface-energy gradient on the substrate or an interfacial-tension gradient on the liquid, able to make the drop moving. In addition, the drop detachment is often favored by a high local temperature that makes both liquid viscosity and surface tension decrease. In the case of a magnetic fluid, the drop manipulation can also be achieved by means of a magnetic field [2–4]. The magnetic liquid is generally a ferrofluid, which is a stable colloidal suspension of nanometer-sized magnetic particles. In the absence of any magnetic field, the behavior of a ferrofluid drop is the same as any regular viscous drop. Under a magnetic field, a ferrofluid drop elongates in the direction of the applied field, offering a straightforward means of manipulating it [5–7].

A particle of ferrofluid in a magnetic field experiences a force per unit volume given by $\overrightarrow{f_m} = \vec{M} \cdot \vec{\nabla} \vec{B}$, where $\vec{M}$ is the magnetization of the fluid and $\vec{B}$ is the applied magnetic flux density [8]. The above equation implies that the applied field is varying much more slowly than the relaxation time for the magnetization ($\sim 10^{-7}$ s), which is in the same direction as the applied field and closely follows the Langevin law for paramagnets [9]. The ferrofluid magnetization is assumed to be a linear function of the magnetic field: $\vec{M} = \frac{\chi_m}{\mu_0 \mu_r} \vec{B}$, where χ_m is the susceptibility and μ_r the relative magnetic permeability of the ferrofluid. The ratio $\chi_m' = \frac{\chi_m}{\mu_r}$ of the magnetic fluid properties is generally introduced to simplify the equations and expresses the balance between the magnetic fluid properties. Note that for low permeability, the susceptibility χ_m' is close to χ_m.

The deformation of a pendant drop under the action of gravity has been extensively studied (see Eggers [10] and references therein). Its equilibrium shape is the result of a competition between gravity, which tends to detach the drop, and the surface tension, which keeps it connected to the tip of the capillary tube. The ratio of these two forces is given by the gravitational Bond number $B_{\text{og}} = \frac{\rho g V}{\gamma \pi R_n}$, where ρ is the liquid density, g the gravity acceleration, γ the fluid surface tension, R_n the radius of the minimum drop cross-section—or drop neck—where the resultant of the surface tension forces is aligned with the vertical direction, and V the volume of fluid below the neck. It is worth noticing that R_n generally depends on the properties of the capillary tube, such as its dimensions and the wettability of the liquid on its material. Since the early work of Tate [11], it is known that the breakup occurs when B_{og} exceeds a critical value on the order of unity [12,13].

For a magnetic liquid, $\vec{B}$ is responsible for additional force $\overrightarrow{F_m}$ acting on the drop in the direction of the magnetic field [14,15]. This force is an increasing function of χ_m. For a given fluid, the drop deformation can be subjected to hysteresis as B is increased and subsequently reduced; the cause of this phenomenon has been explained by an energy argument, which has been used to predict the drop shape [16,17]. In general, the equilibrium shape of a magnetic drop thus results in the competition among magnetic, gravitational, and capillary forces. It therefore depends on two Bond numbers, the gravitational Bond number B_{og} and a magnetic Bond number B_{om}, which compares the magnitude of the magnetic force to that of the capillary force [18]. From a theoretical point of view, the minimization of the free energy should allow one to predict the breakup threshold, which involves the two Bond numbers. Numerical simulations of a sessile drop have pointed out that the two critical Bond numbers seem to be coupled in a complex function, their magnitude increases as χ_m' decreases [19]. In the present case of a pendant drop, the magnetic Bond number can be defined as

$$B_{\text{om}} = \frac{F_m}{\pi R_n \gamma} \propto \frac{\chi_m' V |\vec{\nabla} B^2|}{\mu_0 \pi \gamma R_n}.$$

In this paper, we investigate the possibility of controlling the breakup of a stable pendant ferrofluid drop by the application of an external magnetic field. Experiments with millimeter-sized drops have been performed with two different ferrofluids of contrasting susceptibility in order to understand the effect of the physical parameters on the breakup threshold. We show that this threshold is determined by a single Bond number, which takes into account the combined effects of gravity and magnetism.

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II. MAGNETIC FLUIDS

The two ferrofluids considered here are APG E 18 and EFH1 formulated by Ferrotec® and used as received. They are composed of nanometric magnetic particles suspended in an oil base. Hereafter, they will be denoted by superscripts APG and EHF1. Their density, given by the manufacturer, is $\rho^{\mathrm{APG}} = 970$ and $\rho^{\mathrm{EHF1}} = 1210$ kg/m^3. The surface tensions of the two liquids have been measured with a pendant drop tensiometer (DSA 100 from Krüss). By repeating the measurements 10 times, we found $\gamma^{\mathrm{APG}} = 29.8 \pm 0.1$ and $\gamma^{\mathrm{EHF1}} = 24.7 \pm 0.1$ mJ/m^2. All experiments are carried out at 25°C.

The susceptibility χ'_m of each ferrofluid has been determined experimentally by means of the common U-tube method. A U-shaped tube is filled with one of the fluids. Two electromagnets, facing each other, are located on either side of one arm of the U tube and just below the fluid surface. They are composed of a coil fed by a stabilized dc power supply (ALR3003D by ELC) surrounding a conical iron core. They generate a horizontal magnetic field B perpendicular to the arm under study, which induces a difference of liquid height between the two tube branches: $h = \frac{\chi'_m}{2\mu_0 \rho g} B$. Measuring h for various values of B thus allows the determination of χ'_m.

The magnetic flux generated by each electromagnet has been determined with a GM 07 Gaussmeter. The accuracy of the measurement is mainly limited by the finite size of the sensor and errors in its location; corresponding error bars are systematically presented in graphs. Figure 1 shows the measured values of B against the location z along the axis ($x = 0$, $y = 0$) of one of the electromagnets for two different current intensities (for $I = 0.19$ and $I = 1.67$ A). For the two fluids, it has been checked that the results are not altered by the presence of the U tube filled with ferrofluid on the axis of the electromagnet. For both electromagnets, the measurements are well described by the following empirical relation: $B = f(x,y,z)\mu_0 I^{0.77}$. The deviation from the theoretical relationship derived from the Maxwell-Ampère equation, which is linear with I, mainly results from the nonlinear magnetic properties of the iron core.

Figure 2 presents the liquid-height difference h between the two branches of the U tube versus B. From parabolic fitting,

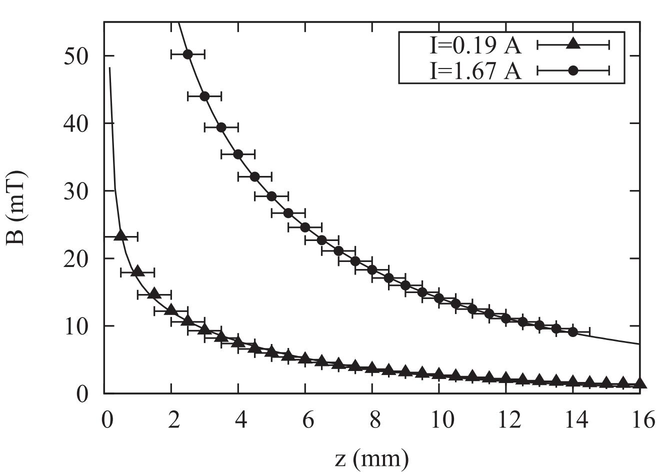

FIG. 1. Magnetic flux B vs. the axial position z. The experimental data are fitted by the empirical relationship $B = f(x,y,z)\mu_0 I^{0.77}$.

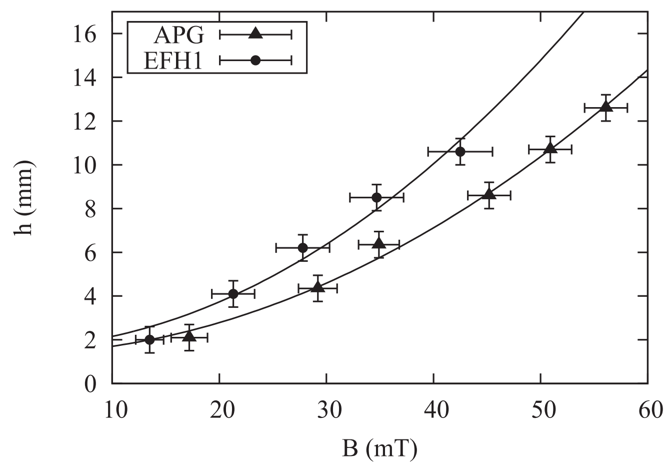

FIG. 2. Difference of liquid height h between to the two U-tube branches vs the magnetic flux B. The experimental data are fitted by the relationship $h = \frac{\chi'_m}{2\mu_0 \rho g} B^2$.

we obtained $\chi'^{\mathrm{APG}}_m = 0.18 \pm 0.01$ and $\chi'^{\mathrm{EHF1}}_m = 0.36 \pm 0.01$. These values are almost at the opposite extremities of the range of commercial ferrofluids. From its low susceptibility, APG is expected to be a linear magnetic liquid whereas EHF1 is likely to be a nonlinear one, i.e. characterized by a susceptibility that depends on the magnitude of B [8,20,21].

III. PENDANT DROP EXPERIMENTS

We have developed specific experimental investigations to study the breakup of a pendant drop under a magnetic field. The motorized injection system and the camera of the tensiometer (DSA 100 from Krüss) are used to generate a pendant drop and take pictures of it (Fig. 3). The drop is formed at the tip of a needle made of polypropylene (Nordson EFD Flexible Green), which has an internal radius $R_{\mathrm{int}} = 0.42$ mm and an external radius $R_{\mathrm{ext}} = 0.60$ mm. Since both of the considered ferrofluids wet the needle material, the interface, where the air, fluid, and tip meet, is located on the needle outer surface.

A preliminary set of experiments has been carried out to determine the breakup threshold under the action of the gravity alone. A small drop is formed and its volume is increased by injecting a small amount of liquid. The radius of the neck of the drop and the volume of fluid V below the neck can

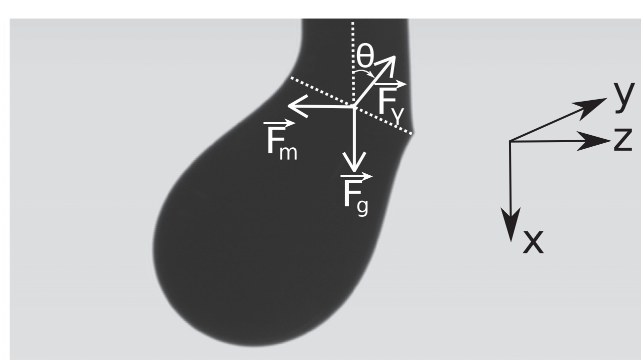

FIG. 3. Picture of a ferrofluid drop (APG) submitted to a magnetic force $\vec{F}_m$. $\vec{F}_\gamma$ is the capillary force and $\vec{F}_g$ is the gravity force.

be measured from the pictures, which allows us to determine the gravitational Bond number. Critical values are obtained from the largest stable drop before breakup under gravity. For both fluids, the critical value of the neck radius R_n is equal to the external radius R_{ext} of the needle. However, since the two fluids have different densities and surface tensions, the critical volumes are not the same. It turns out that the critical volume of the APG drop ($9.0 \pm 0.1\ \mu$L) is larger than that of the EFH1 drop ($6.1 \pm 0.1\ \mu$L), so that the critical Bond number is the same for the two fluids: $B_{\text{ogC}}^{\text{APG}} \approx B_{\text{ogC}}^{\text{EFH1}} = 1.57 \pm 0.02$.

The main set of experiments has been performed in the presence of a magnetic field. One of the two electromagnets used in U-tube experiments is placed in the tensiometer. Its axis is set horizontal and corresponds to the z axis of our Cartesian coordinate frame (Fig. 3). The tip of its conical iron core is located at 5 mm away from the center of the drop in the negative-z side. When an electric current of intensity I passes through the coil, the electromagnet generates, in the vicinity of the drop, a horizontal magnetic field B pointing in the direction of negative z and with a magnitude that decreases with the distance from the core tip. For each value of I, $B(z)$ is calculated from the empirical relationship previously determined (Fig. 1). Assuming a linear magnetic fluid, the magnetic force exerted on the drop can also be calculated by summation over the drop volume: $\overrightarrow{F_m} = \int\int\int_V \frac{\chi_m'}{2\mu_0}\vec{\nabla}B^2 dV$. In the present geometrical configuration, one easily verifies that the z component of $\overrightarrow{F_m}$ dominates the other components (by a factor 20), which can therefore be neglected.

Each experiment is started in the absence of a magnetic field. The drop volume is first adjusted o correspond to the largest stable drop size under the action of gravity. Then, the electric intensity I is increased by steps of 10 mA. The corresponding steps of B change from 0.5 mT for low values of I to 0.2 mT for the large values of I. At each step, a picture of the drop is taken, which allows us to determine R_n and V. The process is continued until the breakup occurs.

Figure 3 displays a typical picture of a pendant drop of ferrofluid strained by a magnetic field. As B is increased, the drop is tilted to a negative angle θ. On the right, the extremity of the capillary tube can be seen, which indicates that the drop is attached to the edge of the tube. On the left, the liquid goes up over to the tube extremity, which confirms that the liquid wets the external wall of the capillary. The way the drop is attached to the capillary can thus evolve as the drop rotates and depends on the wettability of the liquid on the capillary. However, this phenomenon can be ignored when considering the force balance on the volume of liquid below the neck of the drop (dotted line in Fig. 3). This balance involves three forces: $\overrightarrow{F_\gamma} + \overrightarrow{F_m} + \overrightarrow{F_g} = \vec{0}$. The gravity force $\overrightarrow{F_g} = \rho g V \vec{e}_x$ is vertical, the capillary force $\overrightarrow{F_\gamma} = \gamma\pi R_n \overrightarrow{e_\theta}$ is in the direction defined by the tilt angle θ, and the magnetic force $\overrightarrow{F_m} = -\alpha\frac{\chi_m'}{\mu_0} V B^2 \vec{e}_z$ is horizontal. Note that, in the latter expression, B^2 appears instead of $\vec{\nabla}B^2$ because the length scale of the gradient, which does not depend on the magnitude of B, is included in the prefactor α. This static equilibrium yields a simple relationship, $\tan\theta = \frac{F_m}{\rho V g}$, which can be expressed as a function of B: $\tan\theta = \alpha\frac{\chi_m'}{\mu_0 \rho g} B$.

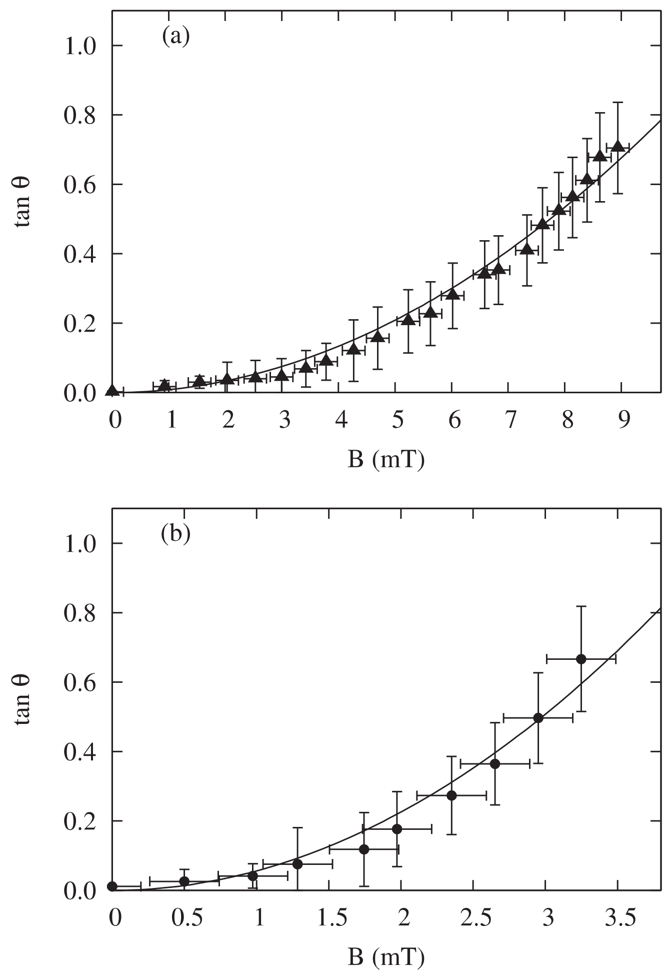

FIG. 4. Tan θ vs the magnetic flux B. (a) APG ferrofluid; (b) EFH1 ferrofluid. The experimental data are fitted by the relationship $\tan\theta = \alpha\frac{\chi_m'}{\mu_0 \rho g} B^2$.

Figure 4 shows the experimental values of B vs. $\tan\theta$ for the two ferrofluids. As expected, a parabolic fitting matches the measurements well. For APG, the fitting gives $\alpha^{\text{APG}} \approx 360\ \text{m}^{-1}$, which is in agreement with the theoretical value computed by summing $\vec{\nabla}B^2$ over the drop volume. On the other hand, for EFH1, the experimental prefactor, $\alpha^{\text{EHF1}} \approx 2300\ \text{m}^{-1}$, is much larger than the theoretical value, $\alpha^{\text{EHF1}} \approx 420\ \text{m}^{-1}$. This discrepancy confirms that EFH1 is a nonlinear magnetic liquid with a large susceptibility [8]. The upper limits in Fig. 4 correspond to the last stable drop deformation before breakup and have been used to estimate the critical values of the magnetic flux, neck radius, and tilt angle, which are given in Table I. The critical magnetic flux is determined within an accuracy of ± 0.1 mT, which corresponds to half the increment between each step at large current values. Since the susceptibility of EFH1 is larger than that of APG, B_c^{EFH1} is smaller than B_c^{APG}. For the same reason, the steps in the tilt angle are larger for EFH1, which results in a lower accuracy in the determination of θ_c for EHF1 than for APG. However, it is remarkable that, within the accuracy of the present measurements, the critical tilt angle is similar for the two ferrofluids: $\theta_c^{\text{APG}} \approx \theta_c^{\text{EFH1}} \approx 35^\circ$. It is also worth mentioning that the reproducibility of the results

TABLE I. Ferrofluids properties and critical values at breakup. ρ is the density, γ the surface tension, χ'_m the susceptibility, V the volume of the pendant drop, θ_c the critical tilt angle of the drop, B_c the critical magnetic flux, and $R_{\rm nc}$ the critical neck radius of the drop.

	ρ (kg/m^3)	γ (mJ/m^2)	χ'_m	V (μL)	θ_c (deg)	B_c (mT)	$R_{\rm nc}$ (mm)
APG	970	29.8 ± 0.1	0.18 ± 0.01	9.0 ± 0.1	35 ± 0.5	9.0 ± 0.1	9 ± 0.1
EFH1	1210	24.7 ± 0.1	0.36 ± 0.01	6.1 ± 0.1	35 ± 3	3.6 ± 0.1	3.2 ± 0.1

has been checked by repeating the experiments and that no hysteresis has been observed when increasing and decreasing the magnetic field below the breakup threshold.

Actually, the breakup threshold depends not only on the magnetic force but also on the gravity force. From the static force balance, it is possible to express the sum of the gravity and the magnetic forces as a function of the tilt angle and the drop weight: $\|\overrightarrow{F_m} + \overrightarrow{F_g}\| = \sqrt{1+\tan^2\theta}\,\rho g V$. We introduce a generalized Bond number accounting for the total destabilizing force as $B_0 = \sqrt{1+\tan^2\theta}\,\frac{\rho V g}{\pi\gamma R_n(\theta)}$.

Figure 5 presents B_0 against the total force $\|\overrightarrow{F_m} + \overrightarrow{F_g}\|$. The two fluids show a similar U curve, starting and ending by the same value of the Bond number. The initial value is obtained in the absence of magnetic field ($B = 0$, $\theta = 0$) and corresponds to the static equilibrium under gravity just below the breakup threshold: $B_0 \approx B_{0gC}$. Then, as the total force is increased, B_0 first decreases, reaches a minimum, and finally rises up until the breakup. The major difference between the two fluids is that since $\rho g V$ is larger for APG than for EFH1, the force has a different behavior along the axis and is dilated for the APG. Since this is the magnetic field which is varied in practice, it is worth mentioning that the final increase of B_0 towards the breakup is as steep for EFH1 as it is for APG when it is plotted as a function of B. In Fig. 5, the final value of B_0 of each curve characterizes the breakup threshold under a magnetic field. It is remarkable, and of major interest for applications, that a single value of the critical Bond number seems to control the breakup of the two ferrofluids, which are contrasted in physical properties: $B_{\rm oC}^{\rm APG} \approx B_{\rm oC}^{\rm EFH1} = 1.57 \pm 0.02$. This suggests that the Bond number based on the resultant of all volume forces exerted on a pendant drop is sufficient to characterize the breakup threshold regardless of the physical nature or the orientation of the applied forces.

To understand why the Bond number first decreases when the total volume force is increased, we have to consider the evolution of the neck radius, which is plotted in Fig. 6. In the absence of a magnetic field, the total volume force is vertical and R_n is equal to the needle radius $R_{\rm ext}$. As F_m is increased, the drop tilts with the total force. Since the liquid wets the needle, R_n is not constrained to remain equal to $R_{\rm ext}$. Indeed, R_n increases to reach a maximum which depends on the drop volume and is found equal to $0.38V^{1/3}$ for both fluids. Hence, when the magnetic field rises, the increase of R_n is faster than the increase of $\|\overrightarrow{F_m} + \overrightarrow{F_g}\|$ and the Bond number therefore decreases. Then, when the increase of R_n slows down, the Bond number reaches a minimum and increases until the breakup threshold. The APG drop being larger than the EFH1 one, R_n has just attained its maximum at the breakup point for the EFH1 fluid, whereas it has already started to decrease for the APG fluid. This particular evolution of the neck radius and the resulting evolution of the Bond number are possible because the total volume force is tilted as its magnitude increases and the liquid wets the needle. If the magnetic force were parallel to gravity, the drop neck could not exceed $R_{\rm ext}$. If the fluid were not wetting the needle, the drop would stay attached to the inner edge of the needle and the drop neck could not become larger than $R_{\rm int}$. In the two latter cases, the Bond number would therefore increase immediately when the magnetic field is applied. In the present situation, since the initial Bond number has been chosen close to its

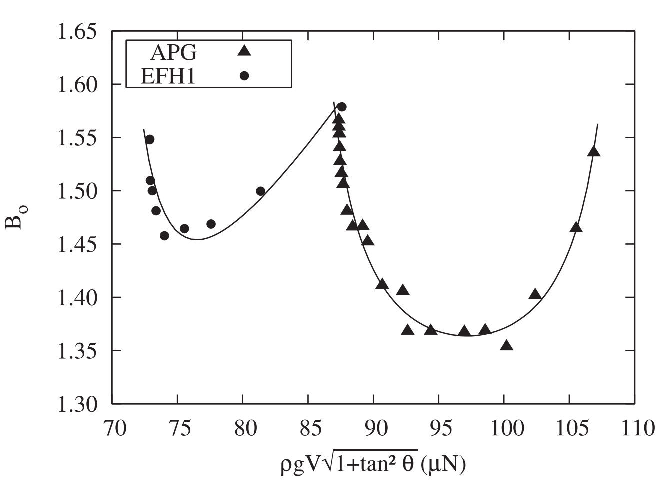

FIG. 5. Bond number B_o vs the total destabilizing force $\|\overrightarrow{F_m} + \overrightarrow{F_g}\| = \sqrt{1+\tan^2\theta}\,\rho g V$, where $\overrightarrow{F_m}$ and $\overrightarrow{F_g}$ are the magnetic and the gravity forces, respectively.

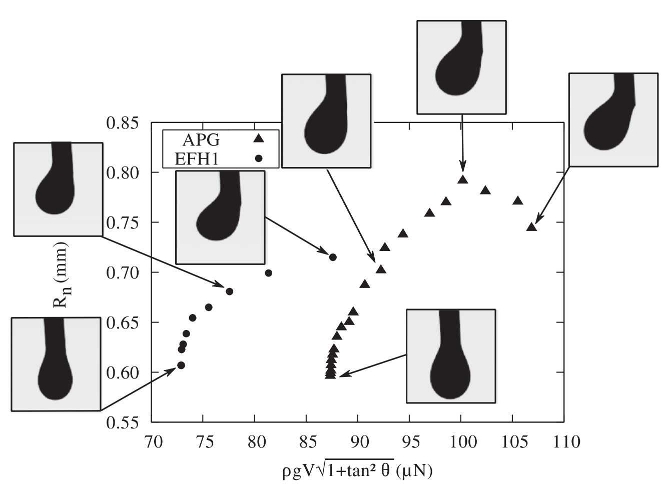

FIG. 6. Neck radius of the drop R_n vs the total force $\|\overrightarrow{F_m} + \overrightarrow{F_g}\| = \sqrt{1+\tan^2\theta}\,\rho g V$, where $\overrightarrow{F_m}$ and $\overrightarrow{F_g}$ are the magnetic and the gravity forces, respectively.

threshold value, the breakup would occur at the first steps of the experiments. In any case, the precise way the breakup threshold is approached seems to have no influence on the critical value of the Bond number.

IV. CONCLUSION

To understand the drop detachment of a liquid under a magnetic field, we have developed specific experiments to study the deformation of a drop by a horizontal magnetic field. Two ferrofluids with different physical properties were used. In a first step, we have determined the critical drop volume at which the breakup occurs under the action of gravity. In these conditions and as expected, the same critical Bond number was obtained for the two ferrofluids: $B_{oC} = 1.57 \pm 0.02$. The drop breakup experiments under a magnetic field were performed with a drop of ferrofluid that has a volume close to the critical one. As the magnetic field is increased, the magnitude of the total volume force exerted on the drop—weight plus magnetic force—increases and its direction tilts relative to the vertical direction. Hence, the drop is deformed and grows in the direction of the total force. Since the liquid wets the needle, the neck of the drop can enlarge up to a certain value that depends on the drop size. This phenomenon allows the resultant of the capillary force to increase and delays the breakup occurrence. Even though we have considered two different magnetic fluids with contrasting properties, the breakup threshold under the magnetic field is determined by a unique critical Bond number, which is similar to that obtained without the magnetic field. These results thus point out that the critical Bond number seems to depend neither on the nature of the force responsible for detachment nor on the complex process that governs the evolution of the drop neck. The critical value found in this work for millimeter-sized systems where gravity plays a significant role is therefore expected to predict the breakup in various applications, including microsystems where electromagnetic forces are predominant.

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