# An Imperfect Subcritical Bifurcation of a Single Ferrofluid Peak

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Here we report on the observation and mechanism of an imperfect subcritical bifurcation that arises when a small amount of ferrofluid is caused to oscillate by a changing magnetic field. Such a bifurcation typically arises in systems due to an imperfection that forces the system in one direction instead of another. We utilize a thermodynamic model and relate this to the fluid's low magnetization susceptibility which is a major factor in causing the observed behavior, providing a physical basis for this bifurcation in the context of a magnetic fluid.

## I. INTRODUCTION

The field of ferrohydrodynamics emerged in the 1960's, motivated by the possibility of converting heat to work with no mechanical parts. Magnetic fluids, or ferrofluids, consist of solid single-domain magnetic nanoparticles 3-15 nm in diameter, and combine the features of magnetism and fluid behavior into one medium. These fluids display novel behavior, most strikingly the spontaneous organization of fluid into geometric patterns of spikes. These patterns, which include hexagons and squares, arise by an impressed magnetic field gradient. This magnetizes the material, leading to a normal field instability manifesting as a series of peaks and valleys. The spin magnetic moments contribute to the molecular field. Spins on neighboring atoms change from parallel to antiparallel alignment, and the electrostatic energy of the system is altered. In some cases, parallel alignment is energetically more favorable, leading to the ferromagnetic behavior, termed paramagnetism, exhibited by iron, nickel, and other materials. Paramagnetism results from the tendency of molecular moments to align with the applied magnetic field but without long-range order, as seen with liquid oxygen and rare-earth salt solutions. A colloidal magnetic fluid (ferrofluid) consists of ferromagnetic single domain particles, leading to superparamagnetism, in which the magnetization in low to moderate fields is much larger compared to paramagnetism. Thus, in a ferrofluid, the domains of a ferromagnetic solid are now free to rotate and translate in solution.

The formation of ferrofluid liquid crests due to magnetic induction was first explored in seminal work by Cowley and Rosensweig in 1967. They provided a theoretical basis for the critical level of magnetization for the onset of the instability at which peaks form and coined this phenomenon the Rosensweig instability. They validated the theory with experimentation by providing a uniform magnetic field, via a Helmholtz coil pair, to a small amount of fluid located between the coils. Oddly, the induced peaks were hexagonal in shape. Measurement of the distances between the peaks showed a close comparison to theoretical predictions. Furthermore, their work showed that the instabilities were governed by the stabilizing gravitational force and interfacial tension, which agreed with established knowledge of these systems. Since then, ferrofluids have proven useful for a range of applications, including the prevention of debris accumulation in hard drives, cooling of speaker coils, and drug delivery. Ferrofluids also provide many ideal characteristics for studying their surface instabilities. One is their colloidal and thermal stability. The magnetic particles remain in suspension, without agglutinating and precipitating out of solution. Thermal energy (Brownian motion) keeps the particles suspended while a surfactant coating on each particle prevents the particles from adhering to each other.

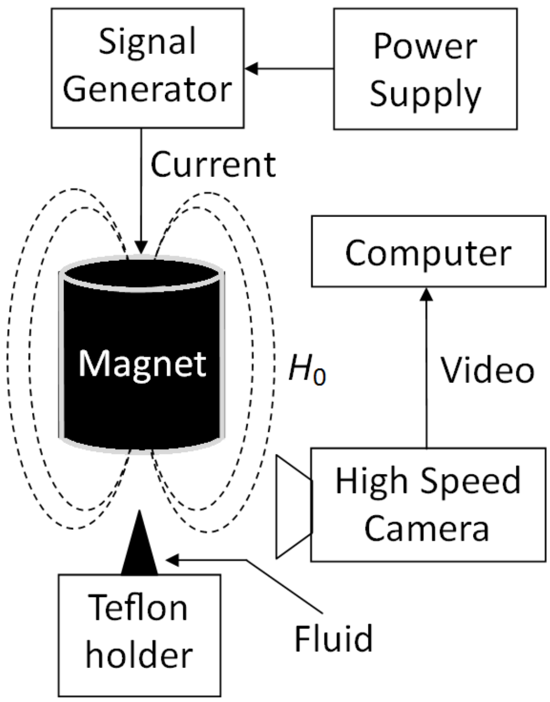


FIG. 1. Experimental setup.

## II. METHODS

The experimental setup, shown in Figure 1, utilizes a round island pole electromagnet (Magnetic Products Inc) to introduce a field instability in the ferrofluid (Ferrotec). The resulting fluid motion is captured on a high-speed video camera (Redlake Motion), and sent to a computer for analysis. Current is sent to the magnet by a power supply (Tecron) and the signal frequency is modulated by a signal generator. The fluid used (EFH-1) has a saturation magnetization ($M_s$) of 44 milliTesla (mT), a dynamic viscosity ($\eta$) of $1.21 \times 10^3$ $A/m$, magnetic susceptibility ($\chi$) of 0.01, a density ($\rho$) of 1240 $kg/m^3$ and surface tension ($\sigma$) of

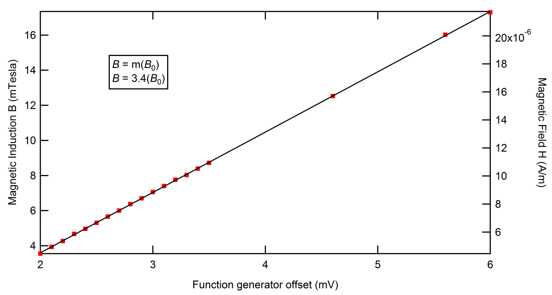


FIG. 2. Magnetic field strength varies linearly with function generator offset.

$1.85{\times}10^-2\ kg/s^2$. The diameter of the teflon fluid vessel was chosen to ensure that its diameter was smaller than the critical wavelength $\lambda_c$ of the normal field instability, where $\lambda_c = 2\pi\sqrt{\sigma/\rho g}$, and $g$ is the acceleration due to gravity (9.8 $m/s^2$). A diameter larger than $\lambda_c$ would give rise to more than one fluid peak under magnetic perturbation. The $\lambda_c$ for our fluid is 7.75 $mm$, while the fluid vessel diameter is 1.6 $mm$. The critical field $H_c$ for the onset of the normal field instability is calculated by Equation 1. For our fluid, $H_c$=$6.96{\times}10^5$ A/m.

$$H_c = \sqrt{\frac{(1+\chi)(2+\chi)2\sqrt{\rho g\sigma}}{\chi^2\mu_o}} \tag{1}$$

While a linear analysis offers simplicity of calculation, a nonlinear analysis is more capable, yielding the amplitude of surface deflections and the character of transitions in peak height. We choose here the nonlinear analysis method, developed by Gailitis[3], in which the total energy $U(z)$ of a magnetically perturbed ferrofluid surface of height $z = z_0(x, y)$ is the sum of the gravitational, surface, and magnetic-field energies, as in equation 2.

$$U(z) = U_g + U_s + U_m \tag{2}$$

Where

$$U_g = \frac{1}{2}\rho g \iint z^2(x, y)dxdy \tag{3}$$

$$U_s = \sigma \iint \left[1 + (\partial z/\partial x)^2 + (\partial z/\partial y)^2\right]^{1/2} dxdy \tag{4}$$

$$U_m = -\frac{1}{2}\iiint \mu_0 M H_0 dxdydz + \frac{1}{2}\iiint \mu_0 {H_0}^2 dxdydz \tag{5}$$

$M$ is the magnetization of the fluid, $H$ is the magnetic field and $\mu_0$ is the permeability constant ($1.257{\times}10^-6 m \cdot kg/s^2 \cdot A^2$). Without a magnetic field ($H$=0), the state's energy minimum is when the fluid is flat ($h$=0). When $H \neq 0$ but is raised beyond $H_c$ while oriented perpendicular to the fluid, the magnetic term dominates and a new energy minimum is born that gives rise to peaks in the fluid. The three equations comprising ( 2) can be further developed to yield an overall thermodynamic potential:

$$\tilde{F} = \rho g \int_{V_{ff}} dV z + \sigma \oint_{S_f} dS + \frac{\mu_0}{2}\int_{V_{ff}} dV H_0\left(r\right) M\left(r\right) \tag{6}$$

At the critical field $H_c$, a hard excitation occurs, characterised by a sudden jump - turning point - in peak height, $z_0 H_c \neq 0$. This supercritical pitchfork bifurcation is not seen in our case. Instead, a soft excitation occurs, characteristic of an imperfect supercritical bifurcation. Such a drastic difference in behavior under similar conditions has been attributed to a difference in magnetic susceptibility between fluids[7], where $\chi$ values greater than $\tilde{5}$ show a sudden turning point[1]. However, recent work has shown the same behavior for $\chi$ values as low as 2.8[6]. Thus, we hypothesize that the current model is incomplete, and utilize the thermodynamic method above.

Here, $V_f f$ is the ferrofluid volume (23.8 $mm^3$) and $S_f$ is the free fluid surface through which the line integral is evaluated. Minimizing $\tilde{F}$ is accomplished using measurements of the free surface profile obtained through video recording of the fluid in motion. Three parameters $h$, $r_1$, $r_2$ and $d_p$ correspond to the peak height, radius of curvature of the peak and valley, and the diameter of the base of the fluid peak, respectively. Thus, $\tilde{F} = \tilde{F}(h, r_1, r_2, d_p)$, and these four hydrodynamic terms are manipulated to minimize this thermodynamic potential, and were measured from when the fluid was at rest ($h$=0) to when it was perturbed to some maximal height before returning to rest again. Surprisingly, the dimensions of fluid only varied over small ranges during this process: $r_1 = 0.3 \pm 0.1mm$, $r_2 = 0.9 \pm 0.2mm$ and $d_p = 3.0 \pm 0.1mm$ and are therefore largely independent of the magnetic field. Thus, the average values were used. This is a noteworthy observation in that we have utilized dynamic models (not shown) from previous work[2654]to analyze the effect on the bifurcation shape between a non-uniform magnetic field by our pole magnet and a uniform magnetic field produced by a Helmholtz coil pair. In both cases, instead of a smooth transition in the peak height, a rapid jump was seen. This lack of correlation between magnetic field strength and both (a) fluid geometry and (b) symmetry breaking behavior indicates that some factor internal to the fluid is causing the smooth bifurcation not seen before. We hypothesize that the very low $\chi$ value of the fluid was the cause for this.

Two types of experiments were conducted. The first used a manually incremented increase in the magnetic field from 9.5 mT up to 19.5 mT and back down. We skipped 0 - 9.5 mT because these field strengths did not produce a peak. This “static ramp” was produced by increasing the voltage signal on the function function generator from 2.8 to 5.6 mV. Next we conducted “dynamic ramps”, where a 1 Hz triangle was was implemented, ramping from 0 mT to 12 mT (3.6V) to zero,

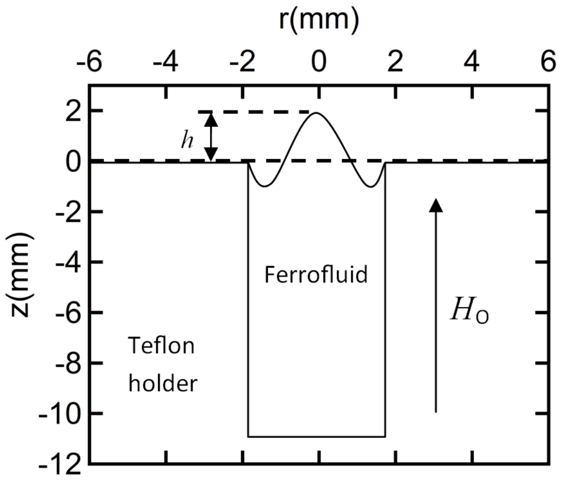


FIG. 3. Teflon vessel with ferrofluid profile under a magnetic field $H_0$ normal to the teflon surface.

and later from 0 mT to 15 mT (5.2V) to zero. Both waves occurred in 0.25 seconds (0.5 Hz). This dynamic behavior was conducted with a high speed camera, then processed with a MatLab script that played back the video while recording the peak height as a function of time.

## III. RESULTS AND DISCUSSION

The ferrofluid instability was very different from anticipated. Figures 4, 5, and 6 show the results of the various perturbations conducted.

In the static case, no jump takes place, but instead we see an imperfect supercritical bifurcation that exhibits no hysteresis. The non-hysteretic behavior is expected, given the slow rate of change of the magnetic field. Several seconds of rest occurred between each

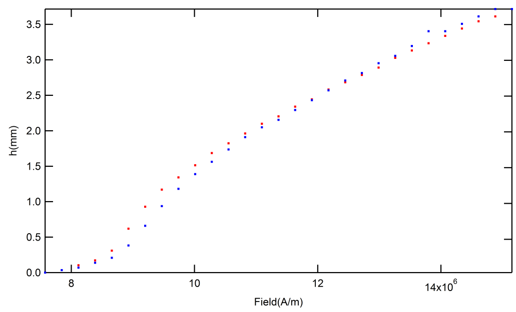


FIG. 4. Fluid height during an upward ramp (red dots) and downward ramp (blue dots) for a statically incremented magnetic field

increment which allowed the fluid to adjust to the previous increment in field strength. In the case of the normal field instability for the two dynamically ramped cases ( 5 and 6), there is obvious hysteresis. The fluid lags behind the triangle wave by 1/8 of a second in the 3.6V and 1/4 of a second in the 5.2V experiment. Surprisingly, the fluid in both cases descends after reaching a maximal peak height bounded by the driving function, only to momentarily slow before continuing on. It is the most pronounced in the 5.2V ramp, in which the fluid almost stops on its way to a maximum height before continuing. Then, in descent, the fluid in fact reverses course by ascending 0.5 mm nearly half-way down to $h$=0. It then descends fully to rest.

We found both the imperfect supercritical bifurcation and the height reversal in the two triangle ramp experiments to be fascinating. However, we only deal with the bifurcation

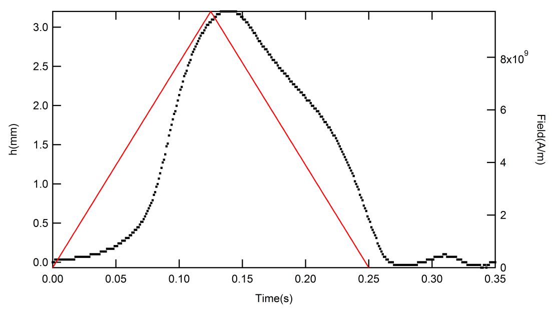


FIG. 5. The driving signal (red lines) set to 0.5Hz with a peak field strength of $10\times10^9$A/m. The fluid peak height (black dots) is traced in time.

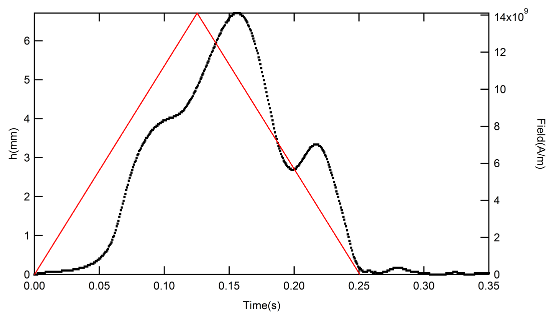


FIG. 6. The driving signal (red lines) set to 0.5Hz with a peak field strength of $14\times10^9$A/m. The fluid peak height (black dots) is traced in time.

here. The three most salient energies that govern a ferrofluid are the hydrostatic, surface, and magnetic-field energy[7]. The governing equations were presented earlier **??**, and were developed to yield a thermodynamic potential. We are hypothesizing that the small $\chi$ value of our fluid is what gives rise to this phenomenon. Early in the experimental process, we were not able to get the fluid to deflect under magnetic fields produced by a Helholtz coil pair nor by placing the teflon block inside of a charged solenoid. After a few iterations, we settled on a large amplifier that supplied the necessary current to a cylindrical pole magnet. The numerical analysis of the thermodynamic model yielded the thermodynamic potentials in 8 and 7. We could apply here the normal form for an imperfect subcritical bifurcation, but such a model is lacking in a physical basis, as it serves only to capture the essence of a variety of systems that undergo such bifurcations. It is apparent from the thermodynamic model that the magnetization susceptibility $\chi$) is embedded in the surface potential term. Here, we hypothesize that the low $\chi$ value of our fluid is responsible for the bifurcation seen, and therefore propose a new normal form ( 7) for the surface energy potential that is specific to this application and is a function of a number of parameters, several of which may be proportional to $\chi$.

$$U_s = a(x-b)^2 + c(x-d)^4 + e. \tag{7}$$

The insight here is that, in the case of the dual minima in 7, an energy "hump" needs to be cleared to go from a peak height of 0 mm to some other peak height sufficiently greater. A small perturbation won't provide

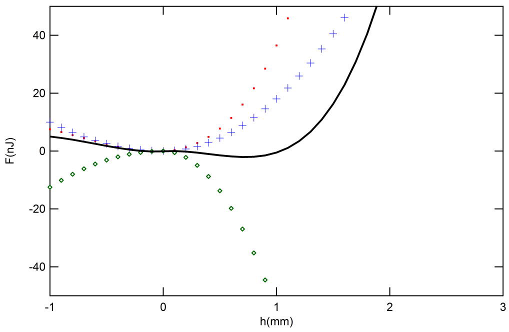


FIG. 7. The hydrostatic energy (blue crosses), surface energy (red boxes), and magnetic energy (green diamonds) are added to give the thermodynamic potential $\tilde{F}$ as a function of peak height $h$. Here, a=1.9, b=d=1.6, c=9, and e=10.5.

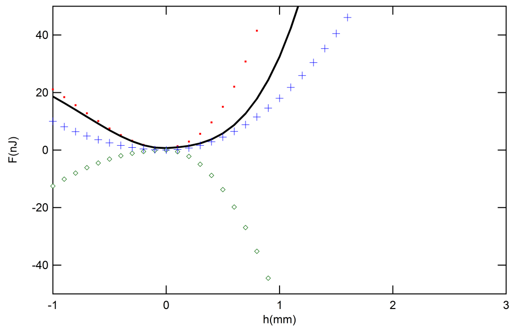


FIG. 8. The hydrostatic energy (blue crosses), surface energy (red boxes), and magnetic energy (green diamonds) are added to give the thermodynamic potential $\tilde{F}$ as a function of peak height $h$. Here, only one thermodynamic minimum is observed near zero peak height. In certain cases, the parameters of ( 7) produce only one minimum. For this case: a=3, b=d=1.85, c=20, and e=34.

sufficient surface energy. This agrees with the nature of the supercritical pitchfork, whereby a substantial magnetic field must develop before the jump can be made, at which poin the fluid rapidly settles into the lower energy well at some nonzero peak height. In the case of 8, the parameters have been adjusted so as to eliminate the second energy minimum. Thus, the fluid will reluctantly proceed up the energy potential, never exhibiting a jump in peak height because there is no other available minimum.

## IV. CONCLUSION

We have reported here for the first time an imperfect supercritical bifurcation in a ferrofluid, as well as a novel set of hysteretic behavior, as seen in the dynamic ramps. These observations and the difficulty in developing a normal field instability in the fluid during experiments led us to hypothesize that the very low $\chi$ value of our fluid was preventing the instability from exhibiting a hard turning point (as has only been observed with ferrofluids to date). We used a thermodynamic model to approximate this behavior, fitting an equation to the surface free energy potential that, leaving the hydrostatic and magnetic potentials constant, were able to vary the equation's parameters to produce one and two energy minima. Two energy minima are indicative of a jump in the peak height, while one minima does not allow for such a jump. With further analysis, it may be possible to give a more physical basis for the terms in the fitted equation, relating them more precisely to the fluid's $\chi$, and bolstering our understanding of this new and interesting behavior.

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