



Experimental investigation on falling ferrofluid droplets in vertical magnetic fields



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ABSTRACT

The dynamic behaviors of a water-based ferrofluid droplet falling in silicone oils subjected to vertical gradient magnetic fields were studied experimentally. The effects of magnetic field, droplet diameter and oil viscosity are investigated. It is found that the droplet undergoes significant deformation with its shape transforming from an initial oblate ellipsoid to a sphere, then to a prolate ellipsoid and finally to a teardrop. We observe more obvious deformation for a bigger droplet in a lower viscosity of the oil and a higher field gradient. Even the satellite droplet appears accompanying with the breakup of the droplet tail, which can be explained by the non-uniform field distribution. To predict the transient velocity of the droplet, a theoretical velocity model is presented. Both the experimental and theoretical results show that the velocity increases with either the field gradient or the droplet size, but decreases as the oil viscosity increases. The behaviors of two falling droplets with different initial diameters in the current magnetic fields were also studied. If the initial separation distance between them does not exceed a threshold, the droplets eventually come into contact to form a dimer moving along the field direction. When the field is suddenly applied in the middle of the journey of the droplet pair, they would be rearranged and aggregated aligning with the field due to the magnetophoretic effects.

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1. Introduction

The study of deformation and breakup of droplets has a long history [1,2]. Unlike rigid particles, a droplet has a deformable interface making its dynamic behavior more complicated. Early experimental and theoretical investigations suggested that when both the droplet and surrounding fluid were perfect dielectric, the droplet would deform into a prolate ellipsoidal shape in a DC electric field [3–7]. Analogous observations have been made when a ferrofluid droplet, in another fluid with different magnetic properties, was exposed to magnetic fields [8,9].

Ferrofluids are stable colloidal suspensions of nanosize magnetic particles in a carrier liquid, such as water or organic oil [10]. The nanoparticles are usually magnetite (Fe_3O_4) and coated by a layer of surfactants to prevent the agglomeration of particles due to the van Waals force. As a class of artificially synthesized fluids with superparamagnetism, ferrofluids combine both the fluidity of fluids and the magnetic properties of solids. These properties make ferrofluid a fascinating material for engineering and biomedical applications. Well-known applications of ferrofluids include their use as liquid seals and bearings in rotating machinery, as dampers in stepper motors and shock absorbers

[11,12]. In medicine fields, ferrofluids are attractive drug carriers due to their abilities to target desired locations with guidance by an external magnetic field, known as magnetic drug delivery [13,14]. Recent progress in synthesis of bio-compatible ferrofluids has made it possible to manipulate and separate live cells in ferrofluid-based microfluidic systems via magnetophoresis [15–18]. Due to the wide applications of ferrofluids, it is both practical and academic interest in understanding the basic mechanisms governing the behavior of ferrofluids under external magnetic fields.

Previous work on ferrofluids is devoted to the equilibrium shape of a free ferrofluid droplet in a uniform magnetic field. The suspended droplet, initially held spherical by surface tension, elongates in the direction of applied field until it reaches a steady shape. It is noted that the influence of magnetic fields on the deformation originates from the Maxwell stress [19]. Since there is a jump in magnetic permeability across the interface, the presence of the droplet alters the magnetic field distribution around it, which in turn affects the Maxwell stress distribution on the droplet surface. This stress depends on both the gradient of permeability and the magnetic field, and can be shown to be largest at the two poles of the droplet, where the axis is the field direction. To maintain the normal stress balance, the curvature increases at the poles and decreases at the equator, leading to the formation of a prolate droplet. Moreover, at a sufficient large value of permeability ratio, the droplet deformation exhibits hysteresis.

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Nomenclature

C_D	drag coefficient	S	maximum horizontal extension of a droplet (m)
Def	deformation factor	T	absolute temperature (K)
d	diameter of a magnetic nanoparticle (m)	t	time (s)
d_{eq}	initial equivalent diameter of a droplet (m)	U_{TS}	terminal velocity without magnetic field (m/s)
d_l	initial equivalent diameter of leading droplet (m)	u	transient velocity (m/s)
d_r	initial equivalent diameter of trailing droplet (m)	u_0	initial velocity at $t = 0$ s (m/s)
E	droplet aspect ratio	V_d	droplet volume (m ³)
Eo	Eötvös number	z	coordinate in the vertical direction
F_m	magnetic force (N)		
F_d	viscous drag force (N)		
g	gravitational acceleration ($g = 9.8$ m/s ²)	<i>Greek letters</i>	
H	applied magnetic field (A/m)	η	viscosity (Pa s)
H_z	z-component of the magnetic field (A/m)	κ	viscosity ratio
H_0	field strength at $z = 4$ mm (A/m)	μ_0	vacuum permeability ($\mu_0 = 4\pi \times 10^{-7}$ H/m)
Δh	initial separation distance of two droplets (m)	ξ	Langevin parameter
K_{vm}	added mass coefficient	ρ	density (kg/m ³)
k_B	Boltzmann constant ($k_B = 1.381 \times 10^{-23}$ J/K)	σ	surface tension (N/m)
L	vertical elongation of a droplet (m)	φ	volume fraction of magnetic nanoparticles
M	ferrofluid magnetization (A/m)		
Mo	Morton number	<i>Subscripts</i>	
M_d	domain magnetization of a magnetic particle ($M_d = 4.46 \times 10^5$ A/m)	c	continuous phase
m_d	droplet mass (kg)	d	dispersed phase
Re	Reynolds number		

Experimental studies on this subject were first reported by Bacri and Salin [9]. They then derived an analytical model which indicated that the droplet aspect ratio at equilibrium was a multiple-valued function of the magnetic field if the permeability ratio above a critical value (21). This hysteresis phenomenon was further numerically confirmed by Lavrova et al. [20,21] using a finite element method, their results were in accordance with the theory of Bacri and Salin. Afkhami et al. [22] developed a volume-of-fluid type code to investigate the equilibrium shapes of a ferrofluid droplet under uniform magnetic fields. It was found that the resulting shape was determined by the balance of magnetic force and interfacial tension.

Since a droplet can serve as an ideal reaction platform or as a vehicle to transport samples in biological and chemical analysis, it is crucial whether a droplet can be easily and precisely manipulated or not. When a ferrofluid droplet placed in a non-magnetic medium is subjected to a non-uniform magnetic field, the Maxwell stress acting on the droplet surface not only deforms the droplet, but also generates a net magnetic force, which causes the droplet to translate. If we consider the ferrofluid droplet as an object with homogeneous magnetic properties, the magnetic force is a body force which is proportional to the gradient of magnetic field and the magnetization of ferrofluids, so the droplet always moves to the region of highest magnetic flux. This feature provides a possibility to remotely control the droplet motion by means of magnetic field gradients. Compared with existing methods for manipulating droplets, such as electrowetting [23], dielectrophoresis [24], and thermocapillarity [25], magnetism has its own advantages, namely, the magnetic force is independent of surface charges, PH, ionic concentration or temperature [26]. Recently, some investigations have been performed focusing on the magnetic actuation of droplets. Guo et al. [27] demonstrated the manipulation of a water-based ferrofluid droplet on a superhydrophobic surface. Rather than using an external permanent magnet, Nguyen et al. [28] controlled the motion of a ferrofluid droplet immersed in silicone oil by using

planar coils. Zakinyan et al. [29] proposed a new approach to transport a ferrofluid droplet with a constant velocity on a solid surface by using rotating magnetic fields. All these experimental results show that the behavior of ferrofluid droplets depends on the magnetic field as well as the flow field. To further reveal the coupling mechanism of magnetic field and flow field, numerical simulations become one of the promising approaches to analyze such issues. Most of the published numerical research on ferrofluid droplets was limited to uniform magnetic fields [19,22,30–32], while there has been little work related to non-uniform magnetic fields. This is because, for the case of uniform magnetic field, the magnitudes of magnetic field at all the boundaries of computational domain are known, which can yield the boundary conditions for solving the governing equation of magnetic field [22]. However, for the case of non-uniform magnetic field, the unknown spatial variation of magnetic field creates great challenges in determining the boundary conditions of magnetic field, and thus hindering the development of numerical model.

As mentioned earlier, the manipulation of ferrofluid droplets is a promising technique for many applications, which can be achieved using magnetic field gradients. Therefore, a comprehensive understanding of the response of a ferrofluid droplet to non-uniform magnetic fields is more significant than that to uniform magnetic fields. However, to our knowledge, there are few studies regarding the dynamics of a ferrofluid droplet under non-uniform magnetic fields, especially numerical simulations. Due to the difficulty in developing a numerical model, in the present work, we reported on an experimental study of the behavior of ferrofluid droplets under the action of magnetic field gradients. For this purpose, an experiment was carried out by injecting a ferrofluid droplet into quiescent silicone oils in external magnetic fields, where the gradients were generated by a solenoid coil. The influences of the field strength, droplet initial diameter and viscosity of the surrounding oil on the shape and velocity of the droplet were studied by processing images taken by a CCD camera. Moreover, a theoretical transient velocity model of the droplet was presented to

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