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Freezing process of ferrofluid droplets: Numerical and scaling analysesWen-Zhen Fang, Hui Zhang , Chao-Yang Zhang, and Chun Yang ^{*}*School of Mechanical and Aerospace Engineering, Nanyang Technological University, Singapore 639798*

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In this study we present numerical and scaling analyses of deformation and freezing processes of ferrofluid droplets under magnetic field effects. A multiphase flow model coupled with an enthalpy-based lattice Boltzmann model is developed to directly simulate the deformation and subsequent freezing processes of a ferrofluid droplet with considerations of both volume expansion and magnetization effects. Meanwhile, analytical models and scaling analyses are derived to reveal how the morphology of the ferrofluid droplet responds to the magnetic field and how the morphology evolution affects the freezing time. We find that the magnetic force induced by magnetic field gradient is much larger than that induced by the magnetization effect, leading to the flattening or elongation of a ferrofluid droplet under magnetic squeeze or lift conditions. The height of the ferrofluid droplet almost linearly decreases (increases) in the low magnetic strength regime for magnetic squeeze (lift) cases, while it follows a nonlinear scaling law under high magnetic squeeze conditions. Besides, the propagation of the freezing front well obeys the scaling law $h \sim t^{0.5}$ for high magnetic squeeze cases, but deviates much from that at the final freezing stage for both magnetic absence and lift cases.

DOI: [10.1103/PhysRevFluids.5.053601](https://doi.org/10.1103/PhysRevFluids.5.053601)**I. INTRODUCTION**

Freezing phenomena of droplets on subcooled surfaces are related to the safety and working performance of many industrial facilities, such as heat exchangers, wind turbine blades, and aircraft [1–3]. In the past few decades, many anti-icing and deicing techniques have been developed to retard ice formation on surfaces. Among them, passive anti-icing approaches, such as the use of superhydrophobic surfaces or patterned surfaces with microstructure or nanostructure, have been reported to be an efficient way to reduce ice accumulation [4–6]. However, such approaches are not tunable once the surfaces are fabricated. Alternatively, the active droplet manipulation strategies using external electric [7] or magnetic fields [8] to manipulate the droplets on surfaces would be a promising strategy to control the freezing process of droplets.

Ferrofluids are colloidal liquids with magnetic nanoparticles dispersed in base fluids (e.g., water or organic solvent). The quick response characteristic of a ferrofluid upon the magnetic field makes it an attractive medium to manipulate droplets. Although many researchers have adopted the active magnetic field to manipulate the mobility and deformability of the ferrofluid droplet [9–12], few studies have reported on how the morphology evolution of ferrofluid droplets affects their subsequent freezing process. In this study, the morphology of ferrofluid droplets is manipulated by imposing an external magnetic field to control their freezing processes. Particularly, the influences of morphology evolution of ferrofluid droplets on the subsequent freezing process will be elucidated.

The shape change of ferrofluid droplets during freezing can be ascribed to two effects: magnetic forces acting on the ferrofluid droplet and volume expansion caused by the phase change from water

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(1000 kg/m³) to ice (916 kg/m³). Without an external magnetic field, the volume expansion plays a dominating role in the droplet morphology evolution [12]. The previous studies have demonstrated that the height increase and the sharp tip formed at the cusp of frozen water droplets are mainly due to the volume expansion effect [13,14]. However, most studies neglect the effect of height increase on freezing time. In this regard, Zhang *et al.* [15] developed a theoretical model to consider the volume expansion effect on the final morphology of droplets, and then adopted the fixed final morphology of droplets to calculate the freezing time by neglecting effects of morphology evolution. Vu *et al.* [16] developed a front-tracking method to investigate the freezing process of droplets by considering the dynamic morphology evolution caused by the volume expansion. However, such model needs two prescribed parameters (trijunction growth angle and contact angle) to close the model. In this study, an improved lattice Boltzmann (LB) model is developed to directly simulate the morphology evolution of ferrofluid droplets with inclusion of the volume expansion effect.

In the presence of a magnetic field, the magnetic force induced by the gradient of the magnetic field and caused by the magnetization effect would lead to the deformation of ferrofluid droplets. How the morphology of ferrofluid droplets responds to the external magnetic field is of importance. Acquiring such understanding using experimental methods alone is nontrivial, and the numerical simulations and scaling analyses would help. In this study, we present both the numerical model and scaling analyses to explore how the morphology of a ferrofluid droplet, especially the droplet height, responds to the magnetic field. With the aid of direct numerical simulations, we can provide detailed morphology evolution of the ferrofluid droplet during the freezing process.

The freezing rate and freezing time of droplets are key issues in ice formation [17]. In previous literature works, the freezing process was approximately treated as a one-dimensional Stefan problem, and a scaling law $h \sim t^{0.5}$ (h is the freezing front height and t is the freezing time) was derived for the prediction of freezing front propagation [18–20]. However, such simplified treatment fails to capture the accelerated propagation of the freezing front at the final freezing stage [21]. Due to the large ratio of thermal conductivity between the ice and surrounding air, most of the heat is transferred through the ice domain, and the shape of the isothermal line is mainly determined by the droplet profile [13,22]. The freezing front is no longer flat at the droplet's upper part, resulting in its deviation from the scaling law $h \sim t^{0.5}$. Thus, a more comprehensive analysis is necessary to predict the propagation of the freezing front at the final freezing stage.

In this study, a multiphase flow LB model coupled with an enthalpy-based thermal LB model is developed to simulate the dynamic freezing process of ferrofluid droplets under magnetic squeeze and lift conditions. The volume expansion and magnetization effects on the evolution of droplet morphologies are included in the models. Meanwhile, the analytical model and scaling analyses are provided to reveal how the magnetic field affects the droplet morphology evolution and subsequent freezing process.

II. NUMERICAL MODEL

In simulations, a multiphase flow model and a heat transfer model are necessary to describe the droplet morphology evolution and the freezing process, respectively. In this section, both the multiphase flow LB model and the thermal LB model are briefly introduced.

A. Multiphase flow

A two-dimensional nine-velocity (D2Q9) LB model is adopted to solve the multiphase flow. The evolution equation of the density distribution function is expressed as [23,24]

$$f_i(\mathbf{x} + \mathbf{c}_i \Delta t, t + \Delta t) - f_i(\mathbf{x}, t) = -\frac{1}{\tau_f} [f_i(\mathbf{x}, t) - f_i^{\text{eq}}(\mathbf{x}, t)] + \Delta t F_i + \Delta t \omega_i S_\rho, \quad (1)$$

where f_i is the density distribution function along the i direction at the space site $\mathbf{x}$ and time t ; and f^{eq} is the equilibrium distribution function given by

$$f_i^{eq} = \omega_i \rho \left[1 + \frac{3}{c^2} (\mathbf{c}_i \cdot \mathbf{u}) + \frac{9}{2c^4} (\mathbf{c}_i \cdot \mathbf{u})^2 - \frac{3}{2c^2} u^2 \right], \quad (2)$$

where ω_i is the weight parameter, $\omega_0 = 4/9$, $\omega_{1-4} = 1/9$, $\omega_{5-8} = 1/36$, and c_i is the discrete velocity,

$$\mathbf{c}_i = \begin{cases} (0, 0)c, & i = 0 \\ (\pm 1, 0)c, (0, \pm 1)c, & i = 1, \dots, 4, \\ (\pm 1, \pm 1)c, & i = 5, \dots, 8 \end{cases} \quad (3)$$

where $c = \Delta x / \Delta t$, in which Δx and Δt are the space step and time step, respectively.

The relation between the kinematic viscosity and relaxation time coefficient (τ_f) is

$$\nu = \frac{1}{3} (\tau_f - 0.5) \frac{\Delta x^2}{\Delta t}. \quad (4)$$

The local macroscopic density and velocity can be determined by

$$\rho = \sum_i f_i, \quad (5)$$

$$\rho \mathbf{u} = \sum_i f_i \mathbf{e}_i + \frac{\Delta t}{2} \mathbf{F}, \quad (6)$$

where $\mathbf{F} = (F_x, F_y)$ is the total force acting on the fluid, including the fluid-fluid interaction force $\mathbf{F}_f$, fluid-solid interaction force $\mathbf{F}_s$, and other body force $\mathbf{F}_b$ (such as the magnetic force $\mathbf{F}_m$ and gravitational force $\mathbf{F}_g$).

The fluid-fluid interaction force can be calculated by [25]

$$\mathbf{F}_f = -G \psi(\mathbf{x}) \sum_i w(|\mathbf{e}_i|^2) \psi(\mathbf{x} + \mathbf{e}_i) \mathbf{e}_i, \quad (7)$$

where G is the interaction strength, and ψ is the pseudopotential function,

$$\psi(\mathbf{x}) = \sqrt{2(p_{\text{EOS}} - \rho c_s^2) / G c^2}, \quad (8)$$

where p_{EOS} is the nonideal equation of state, and the Peng-Robinson equation of state is used,

$$p_{\text{EOS}} = \frac{\rho R T}{1 - b\rho} - \frac{a\varphi(T)}{1 + 2b\rho - b^2\rho^2}, \quad (9)$$

$$\varphi(T) = \left[1 + (0.37464 + 1.54226\omega - 0.26992\omega^2)(1 - \sqrt{T/T_c}) \right]^2,$$

where $a = 0.45724R^2T_c^2/p_c$ and $b = 0.0778RT_c/p_c$. In this work, the saturation temperature is set to be $T_s = 0.85T_c$.

The fluid-solid interaction force can be calculated by

$$\mathbf{F}_s = -G_w \psi(\mathbf{x}) \sum_i w(|\mathbf{e}_i|^2) \psi(\mathbf{x}) s(\mathbf{x} + \mathbf{e}_i) \mathbf{e}_i, \quad (10)$$

where s is the indicator function which is equal to 0 for fluids and 1 for solids, and G_w can be used to vary the wettability of surfaces.

In Eq. (1), the force term F_i can be incorporated by an improved forcing scheme [26,27],

$$F_i = \omega_i \left(1 - \frac{1}{2\tau_f} \right) \left(\frac{\mathbf{c}_i - \mathbf{u}'}{c_s^2} + \frac{\mathbf{c}_i \cdot \mathbf{u}'}{c_s^4} \mathbf{c}_i \right) \cdot \mathbf{F}, \quad (11)$$

where $\mathbf{u}' = \mathbf{u} + \varepsilon \mathbf{F} / (\nu \psi)$ is the modified velocity, and ε is set to be 0.105 in this work.

The magnetic force can be calculated by [28]

$$\mathbf{F}_m = \mu_0 M \nabla H_m, \quad (12)$$

$$M = \varphi M_p L(\xi), \quad (13)$$

$$L(\xi) = \coth \xi - \frac{1}{\xi}, \quad (14)$$

$$\xi = \frac{\pi}{6k_0} \mu_0 M_p H_m d^3 \frac{1}{T}, \quad (15)$$

where μ_0 is the magnetic permeability in vacuum, ∇H_m is the gradient of magnetic field strength, φ is the concentration of Fe_3O_4 particles, M_p is the saturation magnetization of magnetite, k_0 is the Boltzmann constant, T is the temperature of the ferrofluid, and d is the mean diameter of Fe_3O_4 particles.

The gravitational force can be calculated by

$$\mathbf{F}_g = \rho \mathbf{g}. \quad (16)$$

To consider the effect of volume expansion on the droplet morphology evolution, a source term is introduced in the multiphase model [Eq. (1)],

$$S_\rho = \frac{\partial \varphi_s}{\partial t} \rho (1 - \nu), \quad (17)$$

where φ_s is the volume fraction of the ice phase and ν is the density ratio, $\nu = \rho_s / \rho_l$. Physically, the source term should be incorporated into the nodes where solid-liquid phase change occurs. However, the incremental mass at those local nodes cannot immediately propagate towards other liquid nodes since they will be frozen soon. To keep the mass conservation of the entire domain, the source term is evenly divided into those nodes occupied by the liquid phase.

B. Solid-liquid phase change model

To simulate the freezing process of the droplet, a heat transfer equation with solid-liquid phase change should be solved. In the LB model, the evolution equation of the temperature distribution function can be expressed as

$$g_i(\mathbf{x} + \mathbf{c}_i \Delta t, t + \Delta t) - g_i(\mathbf{x}, t) = -\frac{1}{\tau_T} [g_i(\mathbf{x}, t) - g_i^{\text{eq}}(\mathbf{x}, t)] + \Delta t \omega_i S_T, \quad (18)$$

where g_i is the temperature distribution function, and g_i^{eq} is the corresponding equilibrium temperature distribution,

$$g_i^{\text{eq}} = T \left(\omega_i + \frac{1}{2} \mathbf{c}_i \cdot \mathbf{u} \right), \quad (19)$$

where ω_i is the weight parameter

$$\omega_i = \begin{cases} \omega_0 & i = 0 \\ (1 - \omega_0)/4 & i = 1, 2, 3, 4 \end{cases}, \quad (20)$$

and the local temperature can be determined by

$$T = \sum_i g_i, \quad (21)$$

$$a = \frac{1}{2} (1 - \omega_0) \left(\tau_T - \frac{1}{2} \right) \frac{\Delta x^2}{\Delta t}. \quad (22)$$

To consider the solid-liquid phase change issues, the source term in Eq. (18) is introduced as [29,30]

$$S_T = \frac{\varphi_s - \varphi_{s,0}}{\Delta t} \frac{L_f}{c_p}, \quad (23)$$

where φ_s and $\varphi_{s,0}$ are the volume fraction of the solid phase at the current and previous time step, respectively; L_f is the latent heat of the liquid-solid phase change; and c_p is the specific capacity.

In this study, an enthalpy-based method is used to track the solid-liquid interface, in which the local enthalpy is determined by the temperature and solid volume fraction,

$$E = c_{p,w}T(1 - \varphi_s) + L_f(1 - \varphi_s) + c_{p,s}T\varphi_s, \quad (24)$$

where E is the enthalpy. The solid volume fraction can be determined by

$$\varphi_s = \begin{cases} 0 & E < E_s \\ \frac{E - E_s}{E_l - E_s} & E_s \leq E \leq E_l \\ 1 & E > E_l \end{cases}, \quad (25)$$

where E_s and E_l are the enthalpy at the solid and liquid state, respectively.

For the conjugated heat transfer interface located between two domains with different volumetric specific capacity, additional treatments at such interface are necessary to ensure the continuity of the temperature and heat flux. The local interface is assumed to be straight and located halfway between two lattice nodes, and the following treatments are adopted for the conjugated boundaries [31],

$$\begin{aligned} g_{\bar{i}}(\mathbf{x}_l, t + \Delta t) &= \frac{1 - \beta}{1 + \beta} g_i^*(\mathbf{x}_l, t) + \frac{2\beta}{1 + \beta} g_{\bar{i}}^*(\mathbf{x}_s, t), \\ g_i(\mathbf{x}_s, t + \Delta t) &= -\frac{1 - \beta}{1 + \beta} g_{\bar{i}}^*(\mathbf{x}_s, t) + \frac{2}{1 + \beta} g_i^*(\mathbf{x}_l, t), \end{aligned} \quad (26)$$

where $\beta = (\rho c_s)_s / (\rho c_s)_l$, in which the subscripts denote different domains. In Eq. (26), subscript $\bar{i}$ is the opposite direction with respect to i , and the $*$ symbol denotes the distribution function before streaming.

C. Effective properties of ferrofluid droplets and computational domains

In this work, the water-based ferrofluid is considered with a surface tension of 34.8 mN/m. The effective thermal conductivity and diffusivity of the ferrofluid can be determined by the Maxwell model [32],

$$\frac{\phi_e}{\phi_1} = 1 + \frac{3(\alpha - 1)\varphi}{(\alpha + 2) - (\alpha - 1)\varphi}, \quad (27)$$

where φ is the volume fraction of Fe_3O_4 particles; ϕ_e is the effective properties; ϕ_1 is the property of the continuous phase (ice or water); and $\alpha = \phi_f / \phi_1$, in which ϕ_f is the property of Fe_3O_4 particles. The latent heat of the ferrofluid is determined by

$$L_f = (1 - \varphi)L_w, \quad (28)$$

where L_w is the solid-liquid phase change latent heat of pure water.

Initially, a 4.5- μl ferrofluid droplet (radius ~ 1.25 mm) is at rest on a cold copper substrate with its temperature maintained at $T_w = -6$ °C, and the surrounding temperature is 22 °C. The freezing processes of the ferrofluid droplet under magnetic absence, lift, and squeeze conditions are studied. Please see the Supplemental Material [33] for the detailed descriptions of experimental system and methods. Figures 1(a) and 1(b) show the computational domain and external forces acting on the ferrofluid droplet under magnetic squeeze and lift cases, respectively. A 301×301 domain is used

FIG. 1. Schematic of the computational domain and external forces acting on a ferrofluid droplet: (a) the magnetic squeeze condition; (b) the magnetic lift condition.

for the magnetic squeeze cases, while a 201×201 domain is used for the magnetic lift and absence cases, with each grid step representing $25 \mu\text{m}$. These grid systems are chosen on the basis of the simulation results of grid-independence tests for the droplet shape and freezing time.

III. LB MODEL VALIDATION

In this section, two representative benchmarks are simulated to validate the multiphase flow LB model and the solid-liquid phase change LB model. Specifically, we use two cases: (1) the Laplace equation for a suspension droplet and (2) a one-dimensional semi-infinite solidification problem for validations.

For the first benchmark, a suspension droplet surrounded by vapor is located at the center of a 201×201 domain with periodic boundary conditions. The Laplace equation $\Delta P = \sigma/R$, where ΔP is the pressure difference and σ is the surface tension, is tested by varying the droplet radius R . As shown in Fig. 2(a), the pressure drop across the droplet interface linearly increases with $1/R$. The surface tension of the droplet can be determined to be 0.168 (lattice unit) at the saturation temperature $T_s = 0.85T_c$, which agrees well with the results reported in Ref. [34]. Note that the surface tension is related to the saturation temperature [Eq. (9)] and interparticle cohesive force scheme given in the models [Eq. (7)]. The calculated surface tension would help the unit conversions between lattice and physical units, in which the characteristic numbers (Bond numbers) should be equal.

For the second benchmark, the solidification process in a two-region problem (two-phase Stefan problem) is studied. Initially, the entire domain is at a uniform temperature T_i higher than the freezing temperature T_m . Then, the left boundary is subjected to a low temperature T_0 ($T_0 < T_m$). Thus, the solidification starts and propagates along the x positive direction. This is a two-phase Stefan problem since the temperatures in both solid and liquid domain are unknown. The position of the freezing front is [35]

$$x_m(t) = 2\lambda\sqrt{a_s t}, \quad (29)$$

and the temperature distributions in the solid and liquid domain are [35]

$$\frac{T_s(x, t) - T_0}{T_m - T_0} = \frac{\text{erf}[x/(2\sqrt{a_s t})]}{\text{erf}(\lambda)}, \quad (30)$$

FIG. 2. Validations of the accuracy of the multiphase model and the solid-liquid phase change model. (a) Suspension droplet test for the Laplace equation. (b) Solidification in a one-dimensional semi-infinite system. (Here, symbol “Eq. *” denotes Eq. (26). T_i is the initial temperature in liquid phase. $T_i = T_m$ is the one-phase Stefan problem, while $T_i > T_m$ is the two-phase Stefan problem.)

and

$$\frac{T_l(x, t) - T_i}{T_m - T_i} = \frac{\text{erfc}[x/(2\sqrt{a_l t})]}{\text{erfc}(\lambda\sqrt{a_s/a_l})}, \quad (31)$$

respectively. The parameter λ is determined by the following transcendental equation,

$$\frac{e^{-\lambda^2}}{\text{erf}(\lambda)} + \frac{k_l}{k_s} \left(\frac{a_s}{a_l} \right)^{1/2} \frac{T_m - T_i}{T_m - T_0} \frac{e^{-\lambda^2(a_s/a_l)}}{\text{erfc}(\lambda\sqrt{a_s/a_l})} = \frac{\lambda L_f \sqrt{\pi}}{C_s(T_m - T_0)}, \quad (32)$$

where a and k are the diffusivity and thermal conductivity, respectively. If $T_i = T_m$, the solidification process is simplified as a one-phase Stefan problem, and Eq. (32) is reduced to be

$$\frac{e^{-\lambda^2}}{\text{erf}(\lambda)} = \frac{\lambda L_f \sqrt{\pi}}{C_s(T_m - T_0)} = \frac{\lambda \sqrt{\pi}}{\text{St}}. \quad (33)$$

Figure 2(b) shows the comparison of the temperature distribution along the x direction between analytical solutions and the numerical models. In this case, $a_s = 0.005 \text{ m}^2/\text{s}$, $a_l = 0.001 \text{ m}^2/\text{s}$, Stefan number $\text{St} = C_s(T_m - T_0)/L_f = 1$, and $\text{Fo} = a_s t/L_x^2 = 0.625$. In the simulations, the regions where the solid volume fraction $\varphi_s = 0$ and $\varphi_s > 0$ are treated to be liquid and solid nodes, respectively, and the conjugated condition [Eq. (26)] is applied at solid-liquid interfaces. The results show that the numerical results agree well with the analytical solutions only when the conjugated interface [Eq. (26)] is applied. Besides, we find that the freezing front propagation of the one-phase problem ($T_i = T_m = 0^\circ\text{C}$) is faster than that of the two-phase problem ($T_i \neq 0^\circ\text{C}$).

IV. RESULTS AND DISCUSSION

A. Droplet freezing without an external magnetic field

First, the freezing process of the ferrofluid droplet without an external magnetic field is studied. It is well known that the volume expansion during freezing leads to the increase of droplet height [13]. Different from pure water droplets that a sharp tip shows at the cusp when completely frozen, the tip of frozen ferrofluid droplets ($\varphi = 1.8\%$) observed by the experiment [see Fig. 5(e)] is not obvious since its volume expansion is modest ($\rho_f/\rho_s = 1/0.95$), lower than that of pure water ($1/0.917$). Figure 3 shows the increasing normalized height of the frozen droplets h_f/h_d as a function of volume expansion, where h_d is the droplet height before freezing. It is found that the normalized height and

FIG. 3. Variation of the normalized height of frozen droplets (h_f/h_d) with volume expansion (ρ_l/ρ_s). h_d and h_f are the droplet heights before freezing and after frozen, respectively. All the data are calculated without an external magnetic field.

morphology of frozen droplets match well with the experimental results when the volume expansion of the ferrofluid droplet is 1.00/0.95.

The freezing rate of droplets is determined by the local temperature distribution and heat flux at the freezing front. Previous studies have demonstrated that the freezing droplet will experience a rapid recalescence stage, and then followed by a heat transfer dominated freezing propagation. During the recalescence stage, the temperature inside the droplet will restore to the freezing temperature (0°C) [21,36]. However, the initial temperature inside the droplet ($T_{i,d}$) was set to be the freezing temperature [21] or surrounding temperature [22] in the previous numerical studies. Here we study the effect of $T_{i,d}$ on the freezing process of droplets. Figure 4 shows the time-lapsed freezing front propagation (h/R_0) obtained without an external magnetic field. We see that the freezing front propagation at $T_{i,d} = 0^\circ\text{C}$ is faster than that at $T_{i,d} = 22^\circ\text{C}$ since both latent heat and sensible heat are required to be transferred when $T_{i,d} = 22^\circ\text{C}$. Besides, the calculated freezing front propagations at $T_{i,d} = 0^\circ\text{C}$ agree much better with the experimental data than that of $T_{i,d} = 22^\circ\text{C}$. Figure 5 shows the comparison of droplet morphologies and freezing front locations between numerical results ($T_{i,d} = 0^\circ\text{C}$) and the experimental observations, which have a good agreement. Thus, the initial condition $T_{i,d} = 0^\circ\text{C}$ is used hereafter.

In analogy to Eq. (29), the freezing front propagation at the beginning freezing stage follows,

$$h/R_0 = 2\lambda\sqrt{\text{Fo}}, \quad (34)$$

where the Fourier number is $\text{Fo} = a_s t/R_0^2$, in which R_0 is the radius of undeformed droplets, and the parameter λ is the root of Eq. (33) for the one-phase Stefan problem. Note that when $\text{St} = C_s(T_m - T_w)/L_f \ll 1$, it yields

$$\lambda \approx \sqrt{\text{St}/2}. \quad (35)$$

Such scaling law [Eq. (34)] agrees well with both experimental and numerical results (see Fig. 4). Equation (34) also can be derived based on the fact that the freezing front is flat at the beginning stage. The energy conservation equation at the freezing front is given by

$$q_s - q_l = \rho_s L_f \frac{dh}{dt}, \quad (36)$$

FIG. 4. Time-lapsed freezing front propagation (h/R_0) during freezing of ferrofluid droplets without an external magnetic field. R_0 is the radius of the undeformed droplet and $T_{i,d}$ is the initial temperature inside the droplet before freezing. The substrate temperature and environmental temperature are set to be -6°C and 22°C , respectively. Inset shows the temperature variation inside the droplet at $t = 15.5$ s. Symbols “Eq. *” and “Eq. #” denote Eqs. (34) and (38), respectively.

where q_s and q_l are the heat flux at the freezing front computed from the solid side and liquid side, respectively. The temperature distributions along the vertical centerline of the domain and the heat flux $[q/(\rho L_f)]$ at the freezing front are depicted in Figs. 6(a) and 6(b), respectively. It can be seen that the temperature at the unfrozen part of the droplet (behind the freezing front) remains almost 0°C , resulting in an almost zero heat flux q_l at the freezing front. Moreover, at the beginning of the freezing stage, the temperature distribution in the region between the freezing front and the substrate is linear, and thus the heat flux q_s can be calculated by $q_s = k_s(T_m - T_w)/h$ by neglecting

FIG. 5. Comparisons of droplet morphology (solid lines) and freezing front location (dashed lines) between the experimental results (black) and the numerical results (green) during the freezing of ferrofluid droplets at different time moments without an external magnetic field. See the movie in the Supplemental Material [33] for the dynamic freezing front propagation recorded in experiments.

FIG. 6. (a) Temperature distributions along the vertical centerline of the domain. (b) Heat flux at the substrate and the freezing front. (q_s and q_l are the heat flux at the freezing front computed from the solid side and the liquid side, respectively.)

the convection effect [$q_s \sim h^{-1}$, as plotted in Fig. 6(b)]. Thus, Eq. (36) can be rewritten as

$$h/R_0 = \sqrt{2\text{StFo}}. \quad (37)$$

However, the freezing front propagation no longer follows Eq. (34), but speeds up at the final freezing stage. From Fig. 6(a), we can find that the temperature distribution at the region between the freezing front and the substrate is no longer linear at the final freezing stage, and the isothermal lines become concave [see the temperature contour inset in Fig. 6(a)]. Consequently, the heat flux q_s increases at the final freezing stage [see Fig. 6(b)], resulting in acceleration of the freezing front. To derive a scaling analysis, the curved freezing front is assumed to be a portion of a sphere at the final freezing stage (see Fig. S3 in Note 1 of the Supplemental Material [33]). Thus, the heat conduction in this region can be solved in the spherical coordinate, and the freezing front propagation can be expressed as

$$\text{Bo}_s = \mu_0 M^2 R_0 / (2\sigma), \quad (38)$$

$$t'(x) = 2 \frac{k_i(T_m - T_w)t(x)}{\rho_s L_f r_0^2}, \quad (39)$$

where $x = (z_0 + r_0 - h)/r_0$, $\eta = z_0/r_0$, $t'(1)$ is the moment that Eq. (38) is applicable, and z_0 and r_0 are the corresponding location and the radius of the preassumed sphere. The detailed derivation of Eq. (38) is provided in Note 1 in the Supplemental Material [33]. From Fig. 4, we can see that the freezing front propagation at the final freezing stage follows Eq. (38) rather well with $t'(1) = 15.73$ ($t = 14.47\text{s}$) and $\eta = 3.907$. Note that although the simulations are two-dimensional, they can well capture the accelerated freezing front propagation at the final freezing stage.

B. Magnetic squeeze cases

The freezing processes of ferrofluid droplets ($4.5 \mu\text{l}$) under the magnetic squeeze condition are studied in this section. According to Rigoni *et al.* [37], two competing effects, droplet flattening due to the gradient of the magnetic field and droplet elongation due to the magnetization effect, are present when the ferrofluid droplet is subjected to a nonuniform magnetic field. Thus, in addition to the magnetic force induced by the magnetic field gradient [Eq. (12)], another magnetic stress term ($\mu_0 M_n^2/2$) due to the magnetization should be employed at the droplet-air interface to consider the elongation effect [38,39]. To examine the effect of the magnetization induced force on droplet

FIG. 7. Comparisons of the shapes of frozen ferrofluid droplets between the experimental (left side of images) and the numerical (right side of images) results under different magnetic squeeze strengths.

deformation, we perform numerical studies of the deformation of a ferrofluid droplet under uniform magnetic fields [40,41], which shows good agreement with the experimental results [42] (see Note 2 in the Supplemental Material [33] for details),

The morphology of ferrofluid droplets is determined by the interplay between surface tension σ and magnetic forces. In this work, two dimensionless numbers are defined [43],

$$\text{Bo}_m = \mu_0 M |\nabla H_m| R_0^2 / \sigma \approx \mu_0 M |H_m(z_i) - H_m(z_i + R_0)| R_0 / \sigma, \quad (40)$$

$$\text{Bo}_s = \mu_0 M^2 R_0 / (2\sigma), \quad (41)$$

where Bo_m is the magnetic Bond number that denotes the magnetic force caused by the gradient of the magnetic field, while Bo_s represents the magnitude of magnetic stress due to magnetization. By adjusting the magnet distance from the substrate (z_i) under magnetic squeeze conditions, Bo_m varies from 0 to 13.14, while the corresponding Bo_s changes from 0 to 1.23. Hence, the magnetic force caused by the magnetic field gradient is dominating, leading to flattening of the ferrofluid droplets. As shown in Fig. S5 in Note 3 of the Supplemental Material [33], the inclusion of the magnetization effect only leads to slightly more elongation, suggesting that such effect plays a trivial role in the change of droplet morphology. Figure 7 shows the good agreement of droplet morphology between the experimental observations and the numerical results for different magnetic squeeze strength cases.

Figure 8(a) shows the dimensionless height (h_d/h_{d0}) of ferrofluid droplets versus the effective magnetic Bond number ($\text{Bo}_e = \text{Bo}_m - \text{Bo}_s$). Here, h_d is the height of ferrofluid droplets in response to the magnetic field before the onset of freezing and h_{d0} is the height of ferrofluid droplets without

FIG. 8. Effects of magnetic field strength on the morphology of ferrofluid droplets and freezing time under the magnetic squeeze conditions. (a) Deformation of ferrofluid droplets h_d/h_{d0} before the onset of freezing. (b) Dimensionless height (h_f/h_{f0}) of frozen droplets (left axis) and completely freezing time (t_f/t_0) (right axis) versus the effective magnetic Bond number Bo_e . (Symbols “Eq. *”, “Eq. #”, “Eq. &” denote Eqs. (42), (44), and (43), respectively; h_{d0} , h_{f0} , and t_0 are, respectively, the initial droplet height, frozen droplet height, and freezing time without an external magnetic field.)

FIG. 9. (a) Temperature distributions along the vertical centerline of domain at different moments (b) Time evolution of the freezing front under different magnetic squeeze conditions. Symbol “Eq. *” denotes Eq. (34).

an external magnetic field ($\approx R_0$ by neglecting the effect of gravity). It can be seen that the calculated h_d/h_{d0} agrees well with the experimental data. In the low Bo_e regime, h_d/h_{d0} almost decreases linearly with Bo_e , while it follows a $Bo_e^{-0.5}$ relation in the high Bo_e regime. Such relations also can be obtained by the analytical models and scaling analyses. When in the low Bo_e regime, the deformation of ferrofluid droplets is small, and the droplet shape is assumed to be an oblate spheroid. Using a modified Young-Laplace equation [44] and force balance at the pole and equator of the droplet (see Note 4 in the Supplemental Material [33] for the detailed derivation), we have

$$Bo_e = \sqrt{\frac{h_d}{h_{d0}}} + \left(\frac{h_d}{h_{d0}}\right)^{-2.5} - 2\left(\frac{h_d}{h_{d0}}\right)^2. \quad (42)$$

The above analytical model has good agreement with the experimental data within the range $0 \leq Bo_e < 1.5$. When $Bo_e \ll 1$, the droplet is nearly a sphere, and Eq. (42) has an asymptotic solution as

$$\frac{h_d}{h_{d0}} = 1 - 0.25Bo_e, \quad (43)$$

which shows a linear scaling law.

On the other hand, when $Bo_e \gg 1$, the droplet shows a pancake shape, and thus the oblate spheroid assumption no longer holds. Using the modified Young-Laplace equation and a flattened droplet morphology assumption (see Note 4 in the Supplemental Material [33] for details), we have

$$h_d/h_{d0} \approx \sqrt{2}Bo_e^{-0.5}, \quad (44)$$

which can well describe the experimental data [see Fig. 8(a)].

Figure 8(b) shows the dimensionless height (h_f/h_{f0}) of frozen droplets and total freezing time (t_f/t_0) as a function of Bo_e , where h_{f0} is the height of the frozen droplet without an external magnetic field. The ferrofluid droplet experiences expansion during freezing, and its expansion height h_{f0}/h_{d0} is around 1.09 (see Fig. 4) when completely frozen. Hence, h_f/h_{f0} has the same variation trend as the h_d/h_{d0} . As for the total freezing time, we find that it can be precisely predicted with the relation $t_f/t_0 = (h_f/h_{f0})^2$. Since the ferrofluid droplet gets flattened, the isothermal lines inside the droplet are not so concave as those without an external magnetic field. Figure 9(a) shows that the temperature distributions along the vertical centerline of the droplet remain almost linear even at the final freezing stage when $Bo_e = 6.37$. As a result, the propagation of the freezing front follows the relation of Eq. (34) ($h \sim t^{0.5}$) well even at the final freezing stage, as shown in Fig. 9(b). This

FIG. 10. Comparisons of the shapes of ferrofluid droplets before freezing (a), and completely frozen (b), between the experimental (left side of the images) and the numerical (right side of the images) results under different magnetic lift conditions.

is why the predictions of total freezing time with $t_f/t_0 = (h_f/h_{f0})^2$ have a good match with both numerical and experimental results.

C. Magnetic lift cases

The ferrofluid droplet can be elongated by placing the magnet above the substrate. However, the applied magnetic strength cannot be too large so as to avoid possible splitting of ferrofluid droplets. In this section, the freezing processes of the ferrofluid droplets ($4.5 \mu\text{l}$) under the magnetic lift condition are studied within the range $0 < \text{Bo}_e = \text{Bo}_m + \text{Bo}_s < 3.65$. Different from the magnetic squeeze cases, Fe_3O_4 particles dispersed in the droplet can migrate to the unfrozen part induced by the magnetic force during freezing, especially for the high Bo_e cases. To consider this effect, the redistribution of Fe_3O_4 particles is described by assuming the Maxwell-Boltzmann distribution law [43],

$$\varphi = \varphi_0 e^{\mu_0 M H_m \pi d^3 / (6 k_0 T)}, \quad (45)$$

where φ_0 is the particle concentration at the magnetic strength $H_m = 0$. The upper part of the droplet will be further elongated during freezing due to the assembling of Fe_3O_4 particles. Figures 10(a) and 10(b) show the shapes of the ferrofluid droplets before freezing and when completely frozen, respectively. It can be seen that the calculated morphologies of ferrofluid droplets in response to the magnetic field (before the onset of freezing) agree well with the experimental results. However, the final morphologies of the frozen droplet at the high magnetic strength case cannot be precisely predicted by the present numerical model since the detailed migration of Fe_3O_4 particles in ferrofluid droplets is more complicated than the Maxwell-Boltzmann distribution law assumed in the model.

Figure 11(a) shows the dimensionless height (h_d/h_{d0}) of ferrofluid droplets versus the effective magnetic Bond number ($\text{Bo}_e = \text{Bo}_m + \text{Bo}_s$) under the magnetic lift conditions. The calculated h_d/h_{d0} agrees well with the experimental data, and shows an almost linear increment in the studied range of Bo_e . Since the elongation of ferrofluid droplets is modest, especially when $\text{Bo}_e < 1$, we assume that the deformed ferrofluid droplets are prolate spheroid. Using the force balance at the pole and equator of the droplet (see Note 4 in the Supplemental Material [33]), we have

$$\text{Bo}_e = 2 \left(\frac{h_d}{h_{d0}} \right)^2 - \sqrt{\frac{h_d}{h_{d0}}} - \left(\frac{h_d}{h_{d0}} \right)^{-2.5}. \quad (46)$$

As shown in Fig. 11(a), this analytical model agrees well with the experimental data when $\text{Bo}_e < 1$. Primarily because the curvature of the droplet at the droplet-substrate interface no longer obeys the rule of a prolate spheroid [see Fig. 10(a.3)], the predictions obtained by Eq. (46) deviate much from the experimental data when $\text{Bo}_e > 1$. Assuming that the curvature at the droplet-substrate interface has the relation $\kappa = (2 - \omega \text{Bo}_e)/R_0$ (see Note 4 in the Supplemental

FIG. 11. Effects of magnetic field strength on the morphology of ferrofluid droplets and freezing time under the magnetic lift conditions. (a) Deformation of ferrofluid droplet h_d/h_{d0} before the onset of freezing. (b) Dimensionless height (h_f/h_{f0}) of frozen ferrofluid droplets (left axis) and completely freezing time (t_f/t_0) (right axis) versus the effective magnetic Bond number Bo_e . Symbols “Eq. *” and “Eq. #” denote Eqs. (47) and (46), respectively.

Material [33] for details), we have

$$\frac{h_d}{h_{d0}} = \sqrt{1 + \frac{(1 - \omega)}{2} Bo_e}, \quad (47)$$

where ω is constant. As shown in Fig. 8(a), Eq. (47) with $\omega = 0.5$ agrees well with the experimental data.

As discussed in Sec. IV A, the freezing front propagation is accelerated at the final freezing stage due to the convex profile of the droplet. Since the ferrofluid droplet is elongated, the droplet morphology plays a more significant role in freezing front propagation. Figure 12(a) shows the temperature distributions along the vertical centerline of droplets at different time moments. The temperature distribution is no longer linear after $t = 17.80$ s at $Bo_e = 3.65$. As a result, the freezing front propagation significantly deviates from the scaling law $h \sim t^{0.5}$ [Eq. (34)] from that time instant onwards. Figure 12(b) presents the freezing front propagations under different magnetic lift conditions. It can be seen that the deviation from $h \sim t^{0.5}$ increases with Bo_e . This is the reason that

FIG. 12. (a) Temperature distributions along the vertical centerline of the domain at different time instants. (b) Propagations of the freezing front under different magnetic lift conditions. Symbol “Eq. *” denotes Eq. (34).

the plots of $t_f/t_0 = (h_{f,e}/h_0)^2$ in Fig. 11 are significantly different from the experimental results, where $h_{f,e}$ is the height of frozen ferrofluid droplets obtained in experiments.

V. CONCLUSIONS

In the work, we report direct numerical simulations using the lattice Boltzmann method and scaling analyses to provide the underlying physics for the freezing process of ferrofluid droplets under the magnetic squeeze and lift conditions. The morphology evolution of ferrofluid droplets and its influences on the droplet freezing time are two major concerns in our studies.

Under the magnetic squeeze conditions, both the numerical results and scaling analyses show that the height of frozen ferrofluid droplets linearly decreases with the magnetic Bond number (Bo_e) in the low Bo_e regime, but follows a $Bo_e^{-0.5}$ relation in the high Bo_e regime. Under the magnetic lift conditions, the height of frozen ferrofluid droplets almost linearly increases with the magnetic Bond number (Bo_e) in the studied range. The height of ferrofluid droplets is found to significantly affect the droplet freezing time.

The initial temperature inside the droplet ($T_{i,d}$) affects its freezing rate. The calculated freezing time based on $T_{i,d} = 0^\circ\text{C}$ matches the experimental results better than that calculated based on $T_{i,d}$ set to the surrounding temperature. With $T_{i,d} = 0^\circ\text{C}$, the freezing front propagation follows the scaling law $h \sim t^{0.5}$ well at the beginning freezing stage while it deviates much from that during the final freezing stage. Such deviation occurs mainly under the high magnetic lift strength cases while it is significantly reduced under the high magnetic field squeeze cases.

Under the magnetic lift conditions, the particles in the droplet can migrate towards the upper side of the ferrofluid droplet, leading to the further elongation of the droplet during freezing. Under the assumption of the Maxwell-Boltzmann distribution of particles, the calculated droplet morphologies approximately agree with the experimental results. A more detailed model is necessary to consider the migration of particles in a future study, which would help obtain better understanding of dynamic freezing processes.

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