Phase transitions and critical phenomena in the liquid bridge under lateral acceleration

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We study the shape of a liquid body under the influence of a lateral body force in a thin liquid bridge. The shape of the liquid body changes continuously or discontinuously depending on the volume of liquid body. We show that the transition from discontinuous to continuous change is described by the volume-induced phase transition driven by the lateral body force. The order parameter of the phase transition is the amount of shift of the center of mass of liquid body. Critical exponents of a mean-field model have been obtained by numerical calculation. We provide the validity of this critical behavior through the m^4 model of Landau theory of phase transition.

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The investigation of phase transitions and critical phenomena has a long history. This subject has been studied mainly from thermal fluctuations except in the case of quantum fluctuations. We present in this work a phase transition induced by nonthermal fluctuations. The system we treat here is a liquid bridge, a liquid body joining two solid surfaces. The problem of liquid bridges has attracted a great deal of attention in the area of applied science recently [1–3]. For growing single crystals in the floating zone technique [4], liquid bridges can play an important role. In addition, the liquid bridges show an interesting behavior in the change of morphology under the action of a lateral force, which may be described by concepts of conventional critical phenomena and phase transitions.

The energy of a liquid with a free surface is dominated by the surface energy given by the surface tension times the surface area that depends on the amount of deformation and volume. For a liquid bridge under the action of lateral force, the energy of the system is described by the lateral force in addition to those two parameters. The amount of deformation plays the role of order parameter describing the change of morphology of a liquid bridge. Compared with the second order phase transition of the conventional kind [5], one may expect that the lateral force and the volume play the roles of external field and temperature, respectively. We will show that this is the case in what follows. In addition, we analyze the morphology transition phenomena on a macroscopic scale, implying that there are no microscopic fluctuations in the Hamiltonian of a liquid brdige. Thus mean field theory will be sufficient to understand the morphology transition occurring in the liquid bridge. Special features such as thermal fluctuations in nanometer length scale have been reviewed [6].

The experiments on liquid bridges are restricted to special conditions, such as Plateau's tank [7], millimetric liquid bridge cell [8], and spacecraft [4] due to a high sensitivity to body forces. Recently, electromagnetic levitation techniques have been used to compensate the gravity of earth [9]. Many studies have been done on the stability of liquid bridge for various situations of joining solid surfaces and for axisymmetric and nonaxisymmetric liquid bodies [1,10–14]. It has been shown that the shape of a liquid bridge undergoes a discontinuous transition from axisymmetric to nonaxisymmetric as V increases for a given slenderness or as slenderness decreases for a given volume [15,16].

We study the change of the shape of a liquid bridge driven by lateral body forces from the point of view of phase transitions and critical phenomena. Since our interest is not in technical applications but in physical understanding, we consider the simplest possible configuration of a liquid bridge, i.e., a liquid body joining two concentric and parallel disks of the same radius under zero gravity. The inset of Fig. 1 shows the configuration explicitly. This simplest configuration is described by dimensionless parameters, such as slenderness $\Lambda \equiv h/R$, normalized volume $V \equiv V_0/(2h\pi R^2)$, and the Bond number $\eta \equiv \rho a R^2/\nu$, where ρ means the density difference between liquid body and its environment and ν denotes the coefficient of surface tension. V_0 is the volume of a liquid body and a is the lateral acceleration. h and R are geometric scales shown in the inset of Fig. 1.

The purpose of this work is to understand the change of the shape of a liquid body as a volume induced phase tran-

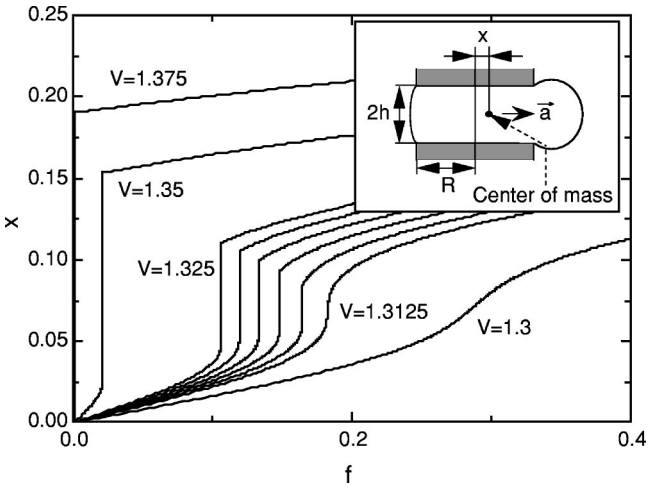

FIG. 1. Change of center of mass X depending on lateral force f for various volumes. The curves between $V=1.3125$ and $V=1.325$ correspond to volumes $V=1.315$, 1.3175, 1.32, and 1.3225, respectively. Inset is the schematic diagram of liquid bridge used in this study.

sition driven by lateral forces. For this purpose, we need to obtain stable equilibrium shape of a liquid bridge under lateral acceleration for a given slenderness. The minimum energy method [14] is a way to get a stable equilibrium shape. The present method finds a stable equilibrium shape of a liquid body by getting a configuration that has equal pressures at any point of liquid surfaces. This method is more effective than the minimum energy method. We cover the liquid surface in terms of finite numbers of triangles for numerical calculations [14,17]. The degree of accuracy increases as the number of triangles increases.

The mechanical pressure is written as

$$P(\vec{r}) = -\frac{\hat{n}_r \cdot \vec{F}_r}{S_r} = \frac{\hat{n}_r \cdot \nabla_r E}{S_r} = \nu \frac{\hat{n}_r \cdot \nabla_r S}{S_r}, \tag{1}$$

where $\hat{n}_r$ is the outward normal unit vector at point $\vec{r}$, $\vec{F}_r = -\nabla_r E = -\nu \nabla_r S$ is the force due to surface tension at point $\vec{r}$ on the surface, and S_r is the small area of surface around the point $\vec{r}$.

The position of the center of mass of a liquid body is obtained after getting a stable shape for a given volume and Bond number and measured from the axis of disks. This plays the role of an order parameter describing the phase transition. Since the quantity $V_0 \eta$ is a measure of lateral body forces for a given liquid and a given geometrical configuration, we introduce a new notation f for this quantity. Figure 1 plots the position of center of mass X expressed in units of R, versus $f \equiv \eta(V_0/R^3) = MaR^2/\nu$ for various volumes when only the value of f increases. As the volume increases, a continuous transition changes into a discontinuous one near a renormalized volume $V_c \approx 1.3125$, above which the liquid bridge has a hump like the one shown in the inset. Experiments [15,16] suggest that the lateral acceleration is most sensitive to symmetry breaking of the shape in the parameter regime $1.3 < V < 1.4$ for the slenderness $\Lambda = 0.225$. We, therefore, choose this region for our theoretical study. Even though many efforts have been devoted to study the effect of lateral acceleration [8,11,12], to our knowledge no analysis has been made from phase transition point of view.

The lines of Fig. 1 are similar to those of the isotherms of a pressure-volume diagram of the liquid-vapor phase transition. This fact leads us to an understanding of the results through an analogy to phase transitions. It is interesting to note that volume V, body force f, and center of mass X in Fig. 1 play the roles of inverse temperature, pressure, and specific volume in the liquid-vapor transition, respectively. There is a hysteresis found in supercooling and supersaturation originating from a surface effect [18]. Therefore, the unstable region [5] expressed by vertical lines in Fig. 1 looks asymmetric. The lines for decreasing f will give another asymmetric unstable region, shifted a little. The Maxwell construction will give rise to a symmetric unstable region, which corresponds to the coexistence regime of left and right humped states of a liquid bridge.

We first analyze the critical behaviors of the second order phase transition. The order parameter ΔX describing second order transition is chosen as the length of vertical straight line for a given $V > V_c$ in Fig. 1. The slopes at broken lines and the line with inflection correspond to compressibilities and the critical isotherm, respectively. We plot these behaviors in Fig. 2.

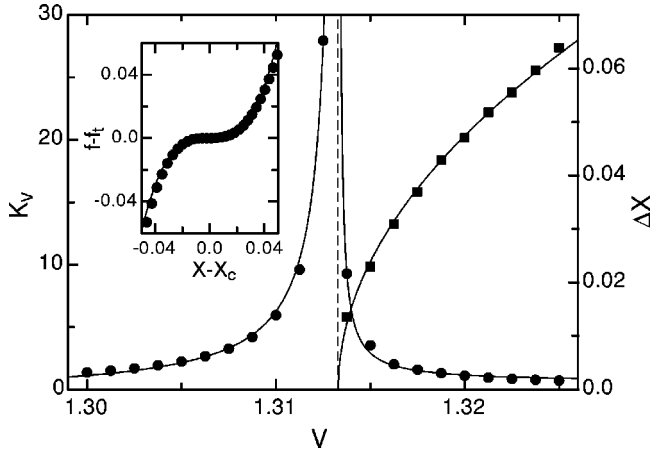

FIG. 2. Fittings for critical exponents β (solid squares), γ's (solid dots), and δ (inset). Solid squares are fitted by $\Delta X \propto |V - 1.3133|^{1/2}$. Vertical dashed line indicates $V_c = 1.3133$. Solid dots are fitted by $K_V \propto |V - V_c|^{-1}$ both for $V < V_c$ and $V > V_c$. Inset is the plot of $f - f_c$ versus $X - X_c$. Fitting curve is $f - f_c \propto |X - X_c|^3$ giving $X_c = 0.0716$.

Solid squares in Fig. 2 show the ordering parameter ΔX versus V. Solid dots show the change of the isochoric compressibility defined by $K_V = (\partial X/\partial f)_V$ by measuring maximum slopes in Fig. 1. The inset of Fig. 2 shows the relation between $f - f_c$ and $|X - X_c|$ at V_c, which corresponds to the equation of states at V_c. Solid lines are least-squares fits for the functions $\Delta X \sim |V_c - V|^{1/2}$, $K_V \sim |V - V_c|^{-1}$, and $f - f_c \sim |X - X_c|^3$ at $V_c = 1.3133$, which is obtained from the first fit. The last fit gives rise to $X_c = 0.0716$. Good least-squares fit in the transition region imply the critical exponent of the order parameter $\beta = 1/2$, compressibility exponents $\gamma = \gamma' = 1$, and the exponent of critical isotherm $\delta = 3$ [18,5]. These are well-known classical values of the exponents. In conclusion, the analysis of Fig. 2 supports a critical transition rather than a crossover.

Even though above numerical analysis strongly implies the existence of critical phenomena in the liquid bridge, theoretical support is needed to clarify it. This can be done by constructing a thermodynamic function appropriate for describing phase transitions. To make the problem simple, we assume a fixed contact line between liquid and solid surfaces, which is the case when the edges are sharp. This condition makes the adhesion energy constant. We also assume the interaction energy inside the surface constant. Thus, the internal energy of this system is governed by the surface energy that is given by free-surface areas multiplied by the surface tension coefficient. Therefore, the internal energy of the liquid bridge system under consideration is controlled by three independent parameters, i.e. volume V, Bond number $2\pi\Lambda\eta$, and the position of center of mass X, for a given geometry and liquid.

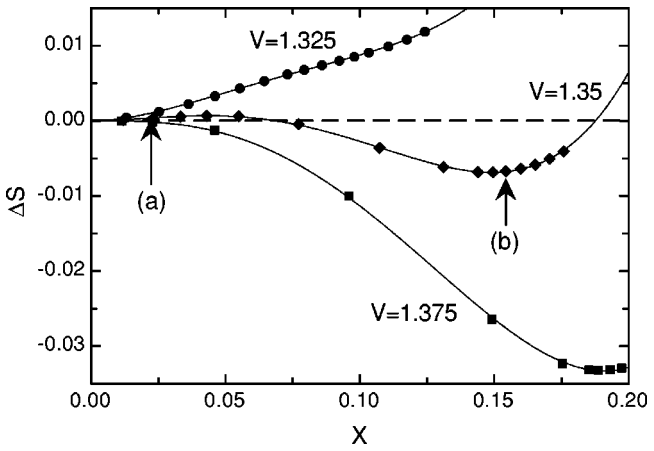

FIG. 3. Changes of surface area depending on X for several V's. Fitting curve is $\Delta S=\frac{1}{2}AX^2+\frac{1}{3}BX^3+\frac{1}{4}CX^4-S_0$, where S_0 is the surface area at $X=0$. Region between (a) and (b) for $V=1.35$ is unstable.

A thermodynamic potential of the liquid bridge for a fixed Λ may be written as

$$\psi(X,V,f)=\nu\oint_{surface}ds-\rho a\int_{volume}rdv$$
$$=S(X,V,f)-fX \tag{2}$$

where s, r, and v denote variables representing surface, lateral distances from the axis, and volume, respectively. In the last expression, S is the area of the free surface of the liquid bridge. Use of R and νR^2 has been made as units of length and energy, respectively.

We expand $S(X,V,f)$ in powers of X. Then Eq. (2) is written as

$$\psi_L(X,V,f)=\frac{1}{2}AX^2+\frac{1}{3}BX^3+\frac{1}{4}CX^4-fX, \tag{3}$$

since the constant term of the expansion for S can be set zero and the linear term vanishes due to the condition of zero force at $X=0$. It is natural to include X^3 in the expansion, because our system is cylindrically symmetric. We will show below the validity of neglecting higher order terms numerically.

The coefficients A, B, and C can be determined numerically by comparing $S(X,V,f)$ with the simulation data. Good agreement between simulation data and surface function shown in Fig. 3 indicates the adequacy of neglecting the terms higher than $O(X^5)$ in Eq. (3). Figure 3 indicates that the area of the surface is decreasing in some region of X as X increases. This region indicated by (a) and (b) for $V=1.35$ corresponds to the unstable region mentioned above.

To describe the critical region, we move the origin of the coordinate of Eq. (3) to (X_c,f_t) in the (X,f) plane in Fig. 1, where f_t denotes the values of f giving a discontinuous transition when $V>V_c$. Moving the origin to (X_c,f_t) corresponds to mapping the liquid-vapor type transition into the

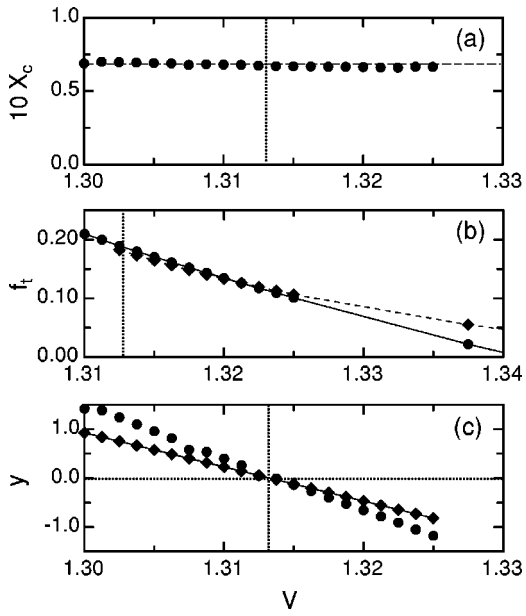

FIG. 4. Check of validity for Eq. (5). Solid dots are simulation data of $-B/3C$ (a), $(A-2CX_c^2)X_c$ (b) where $X_c=-B/3C$, and $(A-3CX_c^2)$ (c). Horizontal dashed line in (a) denotes $X_c=0.0716$. Solid diamonds in (b) are values of f_t and those in (c) denote $y=\alpha|V_c-V|$, where $\alpha=-69.8521$ obtained by β fitting Fig. 2.

magnetic type [19]. The f_t, on the other hand, approaches f_c as $V\to V_c$. Then Eq. (3) can be written as

$$\psi_L(X,V,f)=\frac{1}{2}\alpha(V_c-V)(X-X_c)^2+\frac{1}{4}C(X-X_c)^4$$
$$-(X-X_c)(f-f_t), \tag{4}$$

if the relations $B=-3CX_c$, $f_t=(A-2CX_c^2)X_c$, and $A-3CX_c^2=\alpha(V_c-V)$ are satisfied. Since the constant term of Eq. (4) is physically meaningless, we can neglect it. These three equalities must be checked numerically. The first two relations are satisfied excellently in the transition region. The small difference in the slope of Fig. 4(c) indicates that the α obtained by the simulation data is a little bit different from that through β fitting in Fig. 2. This difference stems from the use of the asymmetric data shown in Fig. 1. We expect very close slopes in Fig. 4(c), if we use symmetric data.

The thermodynamic function of Eq. (4) is just the type of Landau function, in other words the m^4 model, describing phase transitions, which gives the classical exponents of mean-field theory. The liquid bridge we have studied numerically is a classical system treated within mean-field theory, and the transition from continuous to discontinuous changes is of critical behavior, not of crossover.

We close this work with some remarks. We show that the behavior of the change of shape of the liquid bridge can be described by the volume induced phase transition driven by lateral body forces. The critical phenomena of this system follow those of mean-field theory. This is because the energy of the system is expressed by surface tension, which is an averaged macroscopic quantity and therefore does not in-

clude the fluctuation in atomic scale. Having Landau m^4 model (4) from the primitive one (2) provides a theoretical backup for the numerical analysis of this work. There is a correspondence between this system and the magnetic system whose Landau function is given by [18]

$$\psi_L(m,T,H)=\frac{1}{2}b(T-T_c)m^2+\frac{1}{4}cm^4-\frac{mH}{k_BT} \qquad (5)$$

where $b>0$ and $c>0$. The variables, volume V, body force f, and the position of the center of mass X of this work correspond to the temperature $1/(k_BT)$, the external field H, and the order parameter m of the magnetic system, respectively.

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[1] A. S. Andrés, in *Microgravity Fluid Mechanics*, edited by H. J. Rath (Springer-Verlag, Berlin, 1992), pp. 3–17.
[2] J. Meseguer, L.A. Slobozhanin, and J.M. Perales, Adv. Space Res. **16**, 5 (1995).
[3] B.J. Lowry and P.H. Steen, Proc. R. Soc. London, Ser. A **449**, 411 (1995).
[4] I. Martínez, J. M. Perales, and J. Meseguer, in *Proceedings of the Norderney Symposium on Scientific Results of the German Spacelab Mission D-2*, edited by P. R. Sahm, M. H. Keller, and B. Schiewe (Wissenschaftliche Projekfuhrung, Köln, 1995), pp. 220–231.
[5] L. D. Landau and L. P. Pitaevskii, *Statistical Physics*, 3rd ed. (Pergamon Press, New York, 1980), Pt. 1.
[6] L.D. Gelb, K.E. Gubbins, R. Radhakrishinan, and M. Sliwinska-Bartkowiak, Rep. Prog. Phys. **62**, 1573 (1999).
[7] J.A.F. Plateau, *Statique Expérimentale et Théoretique des Liquides* (Gautier-Villars, Paris, 1873).
[8] N.A. Bezdenejnykh, J. Meseguer, and J.M. Perales, Phys. Fluids **11**, 3181 (1999).
[9] M.P. Mahajan, M. Tsige, S. Zhang, J.I.D. Alexander, P.L. Taylor, and C. Rosenblatt, Phys. Rev. Lett. **84**, 338 (2000).
[10] H. Chen and M.Z. Saghir, Microgravity Sci. Technol. **7**, 12 (1994).
[11] J.I.D. Alexander, S. Delafontaine, A. Resnick, and W.C. Carter, Microgravity Sci. Technol. **9**, 193 (1996).
[12] A. Laverón-Simavilla and E. Checa, Phys. Fluids **9**, 817 (1997).
[13] L.A. Slobozhanin and J.I.D. Alexander, Phys. Fluids **10**, 2473 (1998).
[14] F. Zayas, J.I.D. Alexander, J. Meseguer, and J.-F. Ramus, Phys. Fluids **12**, 979 (2000).
[15] M.J. Russo and P.H. Steen, J. Colloid Interface Sci. **113**, 154 (1986).
[16] L.A. Slobozhanin, J.I.D. Alexander, and A.H. Resnick, Phys. Fluids **9**, 1893 (1997).
[17] C. Pozrikidis, *Boundary Integral and Singularity Methods for Linearized Viscous Flow* (Cambridge University Press, New York, 1992).
[18] K. Huang, *Statistical Mechanics*, 2nd ed. (Wiley, New York, 1987).
[19] H. E. Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Oxford University Press, Oxford, 1971).